#### Dyes and Pigments 105 (2014) 105-113



Contents lists available at ScienceDirect

# Dyes and Pigments



journal homepage: www.elsevier.com/locate/dyepig

# Efficient organic light-emitting diodes with low efficiency roll-off at high brightness using iridium emitters based on 2-(4-trifluoromethyl-6-fluoro phenyl)pyridine and tetraphenylimidodiphosphinate derivatives



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## ARTICLE INFO

Article history: Received 10 December 2013 Received in revised form 25 January 2014 Accepted 31 January 2014 Available online 8 February 2014

Keywords: Iridium complex 2-(4-Trifluoromethyl-6-fluorophenyl) pyridine Tetraphenylimido-diphosphinate Organic light emitting diode Efficiency; Efficiency roll-off

# ABSTRACT

Using 2-(4-trifluoromethyl-6-fluorophenyl)pyridine (tfmfppy) as the cyclometalated ligand and tetraphenylimidodiphosphinate (tpip) derivatives as ancillary ligands, three iridium complexes (**1**: Ir(tfmfppy)<sub>2</sub>tpip; **2**: Ir(tfmfppy)<sub>2</sub>ftpip, ftpip = tetra(4-fluorophenyl)imidodiphosphinate; **3**: Ir(tfmfppy)<sub>2</sub>tfmtpip, tfmtpip = tetra(4-trifluoromethylphenyl)imidodiphosphinate) showing phosphorescence at 514, 513 and 508 nm in CH<sub>2</sub>Cl<sub>2</sub> were synthesized, respectively. By using these complexes as emitters, the organic light emitting diodes with the concise configuration of indium tin oxides/1,1-bis(4-(di-*p*-tolyl-amino)phenyl) cyclohexane (60 nm)/**1** or **2** or **3**: bis[3,5-di(9*H*-carbazol-9-yl)phenyl]di-phenyl silane (8 wt%, 30 nm)/ 1,3,5-tri(1-phenyl-1*H*-benzo[d]imidazol-2-yl)phenyl (90 nm)/lithium fluoride (1 nm)/aluminium (100 nm) showed bright green light with maximum luminance higher than 40 000 cd m<sup>-2</sup>. The maximum current efficiency ( $\eta_c$ ) values for **1**, **2** and **3** based devices (**GIr1**, **GIr2** and **GIr3**) were 77.49, 66.57, and 53.26 cd A<sup>-1</sup> at relatively high brightness of 8025, 5286 and 5169 cd m<sup>-2</sup>, respectively. In addition, the efficiency roll-off ratios from the peak efficiency to the brightness of 10 000 cd m<sup>-2</sup> are less than 10% for all the devices. We believe that the good electron mobilities of the phosphorescent emitters lead to the high devices efficiency and low efficiency roll-off. The results suggest that these complexes have potential application in organic light emitting diodes.

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#### 1. Introduction

Phosphorescent iridium complexes play an important part in efficient organic light emitting diodes (OLEDs) due to their high quantum efficiency and short lifetime of triplet excited states [1–16]. The strong spin-orbit coupling (SOC) introduced by the central heavy atom can promote the triplet to singlet radiative transition, resulting unusually high phosphorescence quantum yields at room temperature. On the other hand, since the phosphorescence of iridium complexes primarily originates from the metal-to-ligand charge transfers (MLCT) and the ligand-centered (LC) transitions [17], the energy level of the excited state can be controlled by

\* Corresponding author. *E-mail address:* yxzheng@nju.edu.cn (Y.-X. Zheng). tuning the energy levels of the ligands through substituent effects, which leads to a wide flexible emission color range.

In pursuit of efficient Ir(III) complexes, many heteroleptic complexes, Ir(CN)<sub>2</sub>LX, have been developed, where CN is a monoanionic cyclometalated ligand (*e.g.*, 2-phenylpyridine (ppy), 2-(4trifluoromethylphenyl)pyridine (tfmppy)) and LX stands for a bidentate ancillary ligand (*e.g.*, acetylacetonate (acac), picolinate (pic)) [18,19]. According to the density functional theory calculations, the highest occupied molecular orbital (HOMO) is basically centered on the Ir(III) metal while the lowest unoccupied molecular orbital (LUMO) is generally localized on the cyclometalated ones. Although most ancillary ligands do not make contribution to the lowest excited state directly, they indeed alter the energy levels of the excited states by modifying the electron density at the metal center. Thus, the photophysical property and carrier mobility of iridium complexes can be tuned trough functional substitutes on both cyclometalated and ancillary ligands.

In our previous work, we have reported some high efficient OLEDs based on bis-cyclometalated iridium complexes with tetraphenylimidodiphosphinate (tpip) derivatives as the ancillary ligands [20–24]. The high efficiencies can be explained by more balanced injection and transport of electrons and holes in(to) the emitting layers (EML) due to the good electron mobility of Ir(III) complexes by using the tpip derivatives as ancillary ligands [25]. Therefore, further investigation of the substituted Ir(CN)<sub>2</sub>tpip complexes with new tpip derivatives is possible to prepare more efficient organic light emitting materials and devices.

Fluorination can enhance the electron mobility and result in a better balance of charge injection and transfer, lower vibrational frequency of C-F bond can reduce the rate of radiationless deactivation and the bulky trifluoromethyl substituents can affect the molecular packing and suppress the self-quenching behavior [26]. In this article, a modification was made to ppy by introducing of F/ CF<sub>3</sub> trifluoromethyl groups into phenyl ring at the ortho- and paraposition, as well as each phenyl ring of tpip at the *para*-position. Thus, herein, we report the syntheses, photoluminescence (PL) and electroluminescence (EL) of three novel bis-cyclometalated iridium complexes of  $Ir(tfmfppy)_2$ tpip (1, tfmfppy = 2-(4-trifluoromethyl-6-fluorophenyl)pyridine),  $Ir(tfmfppy)_2 ftpip$  (**2**, ftpip = tetra(4fluorophenyl)imidodiphosphinate) and Ir(tfmfppy)<sub>2</sub>tfmtpip (**3**, tfmtpip = tetra(4-trifluoromethylphenyl)imidodiphosphinate) (Scheme 1).

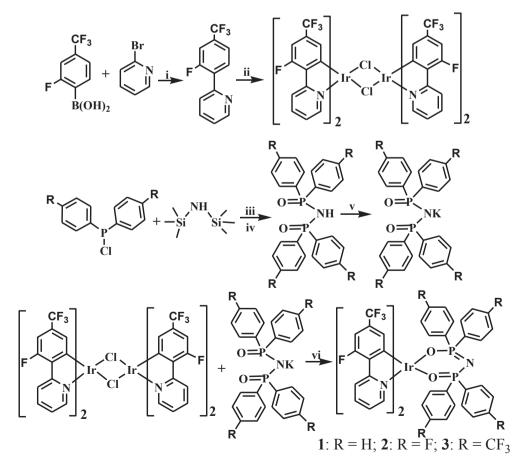
## 2. Experimental section

## 2.1. General information

Commercially available reagents were used without further purification unless otherwise stated. <sup>1</sup>H and <sup>31</sup>P NMR spectra were recorded using a Bruker spectrometer at 500 MHz using TMS as an internal standard. Chemical shifts were recorded by ppm. Multiplicity was denoted by s (singlet), d (doublet), t (triplet), dd (doublet of doublet), m (multiplet). Coupling constants (1) were in hertz (Hz). Mass spectra (MS) were obtained with ESI (Electrospray Ionization, Thermo Fisher Scientific) or MALDI-TOF (Matrix-Assisted Laser Desorption-Ionization Time of Flight, Bruker Daltonic Inc.). Absorption and PL spectra were recorded with a Shimazu UV-3600 and a Hitachi F-4600 spectrometer, respectively. PL lifetime measurements were performed with an Edinburgh FLS-920 spectrometer. The luminescence quantum efficiencies were calculated by comparison of the emission intensities (integrated areas) of a standard aerated aqueous solution of [Ru(bpy)<sub>3</sub>]Cl<sub>2</sub> and the iridium complexes in  $CH_2Cl_2$  solutions according to the equ. (1) [27].

$$\Phi_{\rm unk} = \Phi_{\rm std} \left( \frac{I_{\rm unk}}{I_{\rm std}} \right) \left( \frac{A_{\rm std}}{A_{\rm unk}} \right) \left( \frac{\eta_{\rm unk}}{\eta_{\rm std}} \right)^2 \tag{1}$$

where  $\Phi_{unk}$  and  $\Phi_{std}$  are the luminescence quantum yields of the iridium complexes and  $[Ru(bpy)_3]Cl_2$  solutions, respectively. The  $I_{unk}$  and  $I_{std}$  are the integrated emission intensities of the iridium complexes and  $[Ru(bpy)_3]Cl_2$  solutions, respectively. The  $A_{unk}$  and



Scheme 1. Synthetic routes of ligands and complexes. (i) Pd(PPh<sub>3</sub>)<sub>4</sub>, K<sub>2</sub>CO<sub>3</sub>,THF/H<sub>2</sub>O, 80 °C, 24 h; (ii) lrCl<sub>3</sub>.3H<sub>2</sub>O, 2-EtOCH<sub>2</sub>CH<sub>2</sub>OH/H<sub>2</sub>O, reflux, 24 h; (iii) Anhydrous toluene, 105 °C, 6 h; (iv) H<sub>2</sub>O<sub>2</sub>, THF; (v) KOH, methanol; (vi) 2-EtOCH<sub>2</sub>CH<sub>2</sub>OH, 120 °C, 24 h.

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