

Orange and white electrophosphorescence based on triphenylamine-fluorenyl trifluoromethylpyridine iridium complexes



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ABSTRACT

Novel complexes bis[2-(9,9-bis(4-diphenylaminophenyl)fluoren-2-yl)-5-trifluoromethylpyridinato-C³, N] iridium(acetyl acetonate) (**7**) and bis[2-(9,9-bis(4-diphenylaminophenyl)fluoren-2-yl)-5-trifluoromethylpyridinato-C³, N] iridium (2-picolinic acid) (**8**) were prepared. Trifluoromethyl group and 2-picolinic acid were introduced to tune luminescent emission. Triphenylamine (TPA)-fluorene with large steric hindrance was incorporated to enhance the hole-injection and hole-transport abilities for phosphorescent material, to suppress intermolecular interaction and to shorten fluorescence lifetime. The orange-emissive electrophosphorescent devices for **7** and **8** exhibited stable electroluminescent (EL) peaks at 580 (or 585) and 572 (or 576) nm under a certain concentration at the different bias voltage, and maximum current efficiencies of 32.2 and 35.6 cd/A. The white-emissive devices of the dual emitting layer based on the orange (**7** or **8**) and blue (FIrPic) phosphor showed CIE coordinates of (0.33, 0.41) and (0.31, 0.40), and maximum current efficiencies of 8.9 and 13.8 cd/A. The CIE coordinates with the increasing voltage were stable. High efficiency and slow decay of devices indicated that **7** and **8** were highly efficient orange emitters.

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1. Introduction

White organic light-emitting diodes (WOLEDs) have the promising application in the field of full color display, display backlight, and solid-state light [1,2]. WOLEDs based on phosphorescent materials have significant improvements in efficiency and backlights for full-color active-matrix displays. The most common types of WOLEDs consist of the three separate emitters from red, green and blue [3,4]. To simplify further the device the dual-component orange and blue emitters are doped in two separate layers in an appropriate ratio [5,6]. For example, a 5% orange-emitting iridium complex [Ir(L)₃] (HL=(9,9-diethyl-7-pyridinyl-fluoren-2-yl)diphenylamine) is doped in 4,4'-N,N'-di-carbazolebiphenyl (CBP), and a 8% blue-emitting bis(4,6-difluorophenyl)pyridinato-N,C²) picolinate iridium (FIrPic) in N,N'-dicarbazolyl-3,5-benzene (mCP). The two emitting layers are prepared by co-deposited dopant and host. The

electroluminescence (EL) spectra are strongly dependent on the bias voltage, the color purity is expected to be improved [7,8].

It is crucial that efficient orange-emitting iridium complex phosphors are further explored for the high-efficiency WOLEDs. The electron mobility, thermal stability and volatility for organometallic complex are improved by fluorination [9]. The pyridine ring is modified by a trifluoromethyl group to generate a larger red shift on the emission of the corresponding Ir complexes [10]. To improve high quantum yield and morphological stability and performance of phosphorescent organic light-emitting diodes (OLEDs), it is an important strategy to control host-guest or dopant-dopant interactions effectively. The orthogonally bulky spiro-functionalized (*fac*-Ir(SFP)₃) [11] and spirobifluorene (SPF) and its analogues [12] are introduced. The emission of Ir complex with 2-picolinic acid (picH) (high triplet energy and electron-deficiency structure) in place of acetylacetone (acacH) appears blue-shift and efficient [13]. Furthermore, 9,9-bis(4-diphenylaminophenyl)fluorene offers steric hindrance, high thermal stability and an improved hole injection and transport ability, and suppresses intermolecular aggregation [14].

In our previous work, the trifluoromethylpyridine was introduced to fluorene and spirobifluorene. The ligands

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2-(9,9-diethylfluorene-2-yl)-5-trifluoromethylpyridine (fl-5CF₃-py) and 2-(9,9'-spirobifluorene-2-yl)-5-trifluoromethylpyridine (sfp-5CF₃-py) were synthesized, and iridium complexes (fl-5CF₃-py)₂Ir(acac), (fl-5CF₃-py)₂Ir(pic) [15], (sfp-5CF₃-py)₂Ir(acac) and (sfp-5CF₃-py)₂Ir(pic) [16] were obtained. The orange and white phosphorescent organic light-emitting devices (OLED) were characterized. In this paper, trifluoromethylpyridine and triphenylamine (TPA) were incorporated to fluorene, which enhanced the hole injection and transport for phosphorescent material and suppressed intermolecular interaction. Furthermore, the electron-donating TPA was beneficial to improving the amorphous property of the phosphor emitter and shortening fluorescence lifetime [7]. A novel ligand 2-(9,9-bis(4-diphenylaminophenyl)fluorene-2-yl)-5-trifluoromethylpyridine (pafp-5CF₃-py), two iridium complexes (pafp-5CF₃-py)₂Ir(acac) (**7**) and (pafp-5CF₃-py)₂Ir(pic) (**8**) were prepared. The orange and white OLEDs based on **7** and **8** were fabricated and investigated.

2. Experimental

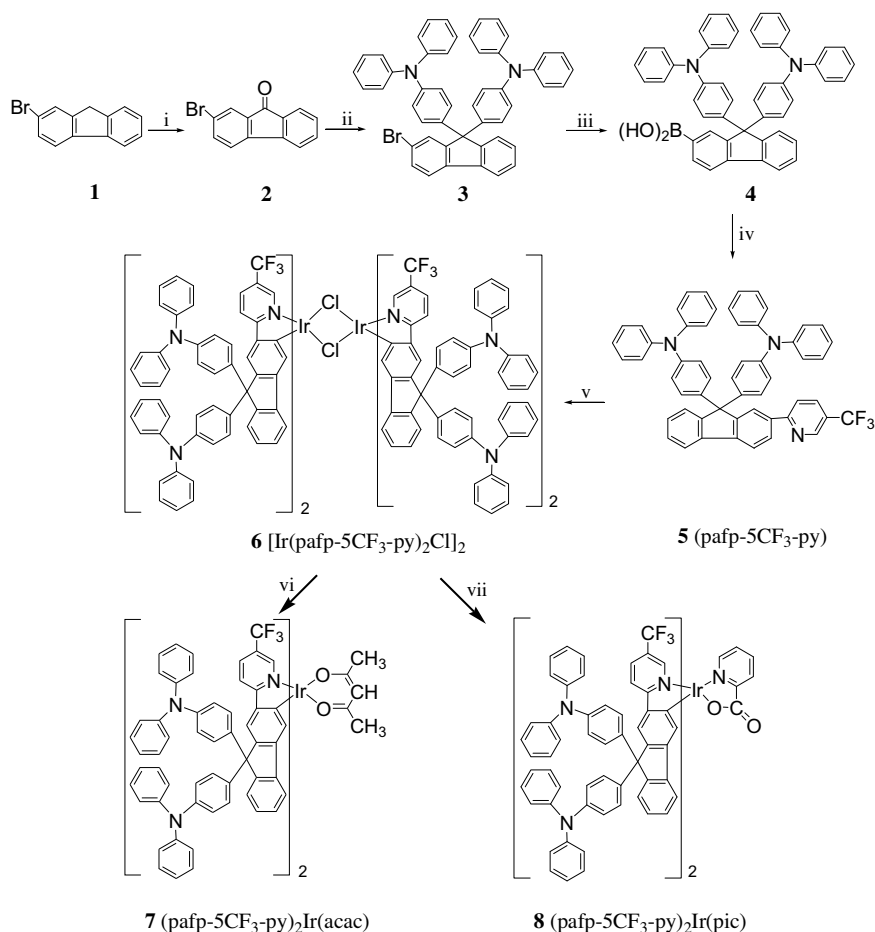
2.1. Materials and instruments

Pd(OAc)₂, Cs₂CO₃, PPh₃, fluorene, 2-chloro-5-trifluoromethylpyridine, *n*-butyllithium (1.6 mol/L), IrCl₃·*n*H₂O and 2-picolinic acid were purchased from Acros Organics. Chemicals and solvents were used as received in the highest commercially available grade unless described otherwise. Tetrahydrofuran (THF) and dioxane

were refluxed with sodium and benzophenone, and purified with distillation. 2-Methoxyethanol was purified by distillation at a reduced pressure. The synthesized compounds were characterized using ¹H and ¹³C NMR spectroscopy (Bruker Avance 400), mass spectrometry analysis (Shimadzu LC-MS2010A), and elemental analysis (Carlo Erba 1106), respectively. Cyclic voltammetry (CV) analyses were performed by using Zahner Zennium electrochemical workstation. Tetra-*n*-butylammonium hexafluorophosphate (TBAPF₆) (0.1 M) was used as supporting electrolyte in an anhydrous dichloromethane solution. An Hg/Hg₂Cl₂ electrode was chosen as the reference electrode, a platinum wire as the counter electrode and a glass carbon rod as the working electrode. Ferrocene was set as a reference material at a scan rate of 50 mV/s. The UV-vis absorption spectrum was recorded by using GBC Cintra 303 Spectrometer. Photoluminescent (PL) spectrum was measured with Perkin Elmer LS-55 luminescence spectrometer. Phosphorescence quantum yield (Φ) was tested against the reference *fac*-Ir(ppy)₃ (Φ_{fac-Ir(ppy)₃} = 0.4) in Ar-saturated toluene (10⁻⁵ M) at 298 K. Phosphorescence lifetime was carried out by Edinburgh Instrument (FLS920) with a laser at the wavelength of 405 nm as an excitation source.

2.2. Synthetic procedures

2-Bromofluorene (**1**) [17] and 2-bromofluorenone (**2**) [18,19] were prepared as described in the corresponding references, respectively.



Scheme 1. A schematic diagram for chemical structures of intermediates, [Ir(pafp-5CF₃-py)₂Cl]₂, (pafp-5CF₃-py)₂Ir(acac) and (pafp-5CF₃-py)₂Ir(pic). Reagents and conditions are: (i) CrO₃, (CH₃CO)₂O, ice water bath; (ii) Triphenylamine, methanesulfonic acid, 140 °C, 6 h; (iii) *n*-BuLi, THF, -78 °C, 1 h; B(OCH₂CH₃)₃, -78 °C, 1 h; rt, overnight; 2 M HCl; (iv) 2-Chloro-5-trifluoromethylpyridine, Pd(OAc)₂, PPh₃, Cs₂CO₃, dioxane, 80 °C, 12 h; (v) IrCl₃·*n*H₂O, 2-methoxyethanol/H₂O (v/v, 3/1), reflux, 24 h; (vi) Acetylacetonate (acacH), Na₂CO₃, reflux, 16 h; (vii) 2-Picolinic acid (picH), Na₂CO₃, reflux, 20 h.

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