



Single emissive layer white phosphorescent organic light-emitting diodes based on solution-processed iridium complexes



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ABSTRACT

We fabricated efficient, white phosphorescent organic light-emitting diodes (WPhOLEDs) with all of blue, yellow, and deep-red iridium(III) complexes doped in 4,4',4''-tris(carbazol-9-yl)triphenylamine [or 1,3-bis(carbazol-9-yl)benzene]:poly(9-vinylcarbazole):1,3-bis[2-(4-*tert*-butylphenyl)-1,3,4-oxadiazol-5-yl] mixed-host system and investigated their photophysical, electrochemical, and electroluminescent properties. An optimal combination of blue, yellow, and deep-red iridium(III) complexes improved the charge balance and triplet excitons confinement in the emitting layer. We fabricated the WPhOLEDs with a configuration of indium tin oxide/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (40 nm)/emitting layer (50–60 nm)/1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (20 nm)/lithium fluoride (1 nm)/aluminum (150 nm). In terms of performance, the WPhOLEDs exhibited a maximum luminance efficiency of 23.04 cd/A and external quantum efficiency of 11.45%.

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1. Introduction

White organic light-emitting diodes (WOLEDs) have been the subject of the most extensive research among full-color displays with color filters, as backlight units for liquid crystal displays and even in lighting applications [1–3]. In order to generate the desired white light emission, mixtures of the three primary colors, red, green, and blue, or two complementary colors, blue and yellow, are typically required, and various approaches for the fabrication of WOLEDs have been reported [4,5]. Most studies have focused on improving the quantum efficiency of WOLEDs: WOLEDs based on the development of fluorescent materials are more mature and the device structures based on them are comparatively simple, but they suffer lower efficiency. In particular, due to the realization of 100% internal quantum efficiency, phosphorescent emitting materials have a strong potential to become more efficient than fluorescent emitting materials [6,7]. Hence, highly efficient WOLEDs are based on phosphorescent emitters such as iridium (Ir) complexes [8–14]. Indeed, white phosphorescent OLEDs (WPhOLEDs) have been

prepared through the doping of Ir(III) complexes into polymeric hosts such as poly(N-vinylcarbazole) (PVK) [15–17].

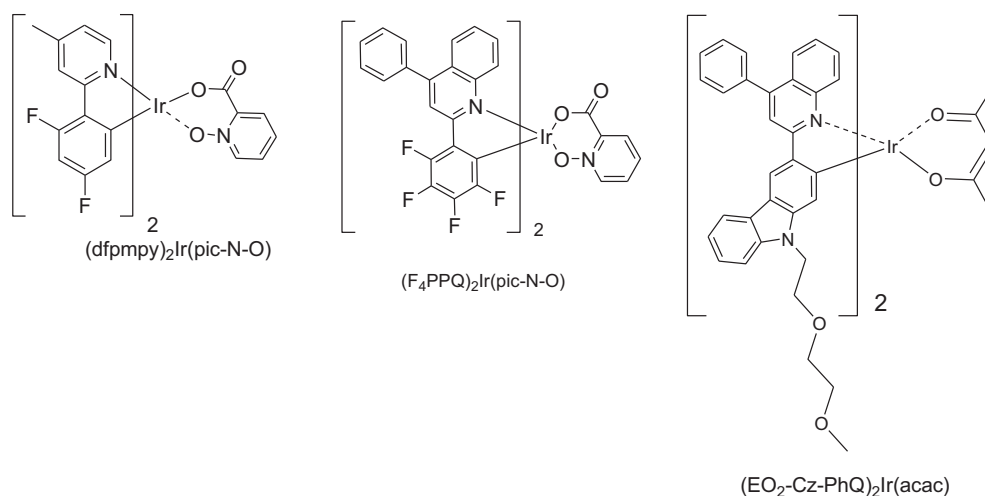
The two fabricating methods for OLEDs are thermal vacuum evaporation for the fabrication of small molecules based on OLEDs and solution processing for those based on oligomers or π -conjugated polymers [18,19], and more recently dendrimers [20,21]. Vacuum deposition enables complicated multilayer device architectures and affords excellent devices with high efficiencies. However, it is difficult to control the location of exciton recombination and the doping concentration in vacuum deposition. On the other hand, multilayer devices cannot be fabricated via the solution processed method because the solvent, which is used for dissolving organic materials, can undergo swelling, leading to damage to the previous layer. However, solution-based processing of organic materials is another attractive technique for low-cost manufacturing devices because it facilitates the codoping of several dopants and provides compatibility with low-cost, large-area manufacture technology such as screen printing and inkjet printing [22]. Thus, WOLEDs offer the great advantage of simplifying fabrication solution processing.

Recently, we developed for the first time highly efficient Ir(III) complexes using picolinic acid N-oxide (pic-N-O) as an ancillary ligand for the fabrication of PhOLEDs. These new Ir(III) complexes

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Scheme 1. Molecular structures of the Ir(III) complex used in this study.

improved the performance of deep-blue, yellow, and red PhOLEDs due to the synergistic effect of both phenylpyridine (ppy) and phenylquinoline (PhQ) based on the main ligand and the pic-N-O ancillary ligand [23–26].

In this work, we report efficient WPhOLEDs with good color purity and color rendering index (CRI) using three phosphorescent Ir(III) complexes doped on a single emissive layer that can be fabricated by solution processing method. We designed and synthesized blue, yellow, and deep-red Ir(III) complexes such as bis[2-(2,4-difluorophenyl)-4-methylpyridine]iridium(III) picolinic acid N-oxide [(dfpmpy)₂Ir(pic-N-O)], bis[4-phenyl-2-(2,3,4,5-tetrafluorophenyl)quinoline]iridium(III) picolinic acid N-oxide [(F₄PPQ)₂Ir(pic-N-O)], and bis[9-(2-(2-methoxyethoxy)ethyl)-3-(4-phenylquinolin-2-yl)-9H-carbazole]iridium(III) acetylacetonate [(EO₂-Cz-PhQ)₂Ir(acac)], respectively, as emitters [24–26]. The single emitting layer (EML) consists of a blend of PVK, 4,4',4'-tris(carbazol-9-yl)triphenylamine (TCTA), and 1,3-bis[2-(4-*tert*-butylphenyl)-1,3,4-oxadiazol-5-yl] (OXD-7) doped with (dfpmpy)₂Ir(pic-N-O), (F₄PPQ)₂Ir(pic-N-O), and (EO₂-Cz-PhQ)₂Ir(acac). In addition, high triplet state and hole transport host, 1,3-bis(carbazol-9-yl)benzene (mCP) was used to prohibit triplet exciton diffusion out of the EML instead of TCTA. Carefully managing the triplet transfer and charge transport via the introduction of such multifunctional materials and smart device engineering are essential for attaining such device performance. Consequently, high performance WPhOLEDs were fabricated with a luminance efficiency of 23.04 cd/A, an external quantum efficiency (EQE) of 11.45% and balanced white light with Commission Internationale de l'Éclairage (CIE) coordinates of (0.326, 0.385), which were close to the ideal CIE coordinates of (0.333, 0.333) for pure white light.

2. Experimental

2.1. General information

All chemicals and reagents were purchased from Aldrich Chemical Co. and used without further purification. The THF solvent was dried and purified by distillation over sodium/benzophenone with N₂ atmosphere and other solvents were used without additional purification. The [(dfpmpy)₂Ir(pic-N-O)], [(F₄PPQ)₂Ir(pic-N-O)], and (EO₂-Cz-PhQ)₂Ir(acac) dopants were synthesized according to the methods in the literature [24–26].

The ¹H and ¹³C NMR spectra were recorded on a Varian Mercury Plus 300 MHz spectrometer in CDCl₃ using tetramethylsilane as an internal reference. The UV–visible and the fluorescence spectra were recorded with JASCO V-570 and Hitachi F-4500 fluorescence spectrophotometers at room temperature. Thermal analyses were carried out on a Mettler Toledo TGA/SDTA 851e and DSC 822e analyzer under N₂ atmosphere at a heating rate of 10 °C/min. Cyclic voltammetry (CV) studies were carried out with a CHI 600C potentiostat (CH Instruments) at a scan rate of 100 mV/s in a 0.1 M solution of tetra-*n*-butylammonium hexafluorophosphate (TBAPF₆) in anhydrous acetonitrile/benzene (1:1.5 v/v). A platinum wire was used as the counter electrode and an Ag/AgNO₃ electrode as the reference electrode. All of the electrochemical experimental studies were carried out in the open air at room temperature.

2.2. Fabrication of WPhOLEDs and measurements

The sheet resistivity and thickness of the indium tin oxide (ITO) thin films coated on glass substrates used in this study were 20 Ω/

Table 1
Photophysical, electrochemical and thermal data for the Ir(III) complexes.

Ir(III) complex	<i>T</i> _d [°C] ^a	<i>T</i> _g [°C] ^b	λ _{abs} [log ε (nm)] ^c	λ _{em} [nm] ^d	λ _{em} [nm] ^e	Φ _{pl} [%] ^f	HOMO/LUMO [eV] ^g
(dfpmpy) ₂ Ir(pic-N-O)	354	193	282(4.61), 376(3.91), 452(1.62)	465, 492	464.490	0.21	−5.59/−2.68
(F ₄ PPQ) ₂ Ir(pic-N-O)	364	245	280(4.94), 349(4.63), 467(3.97)	568	565	0.83	−5.45/−2.89
(EO ₂ -Cz-PhQ) ₂ Ir(acac)	344	150	317(5.10), 378(5.00), 496(4.10)	624	610	0.03	−4.96/−3.06

^a Temperature with 5% mass loss measure by TGA with a heating rate of 10 °C/min under N₂.

^b Glass transition temperature, determined by DSC with a heating rate of 10 °C/min under N₂.

^c Measured in CHCl₃ solution at 1 × 10^{−5} M concentration.

^d Maximum emission wavelength, measured in CHCl₃ solution at 1 × 10^{−5} M concentration.

^e Maximum emission wavelength, measured in film state onto quartz glass.

^f Measured in 1 × 10^{−5} M in CHCl₃ solution relative to Ir(piq)₂(acac) (Φ_{pl} = 0.2) and Irpic (Φ_{pl} = 0.42).

^g Determined from the onset of CV oxidation and UV–visible absorption edge.

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