

Wide bandgap iridium complexes varying by their ancillary ligands: Influence on their electroluminescence properties



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ABSTRACT

Three heteroleptic iridium(III) complexes comprising 1-methyl-3-propyl-5-(2,4,5-trifluorophenyl)-1*H*-1,2,4-triazole (tfpt) as the cyclometalated ligand and varying by their ancillary ligands were examined as triplet emitters for Phosphorescent OLEDs. Interestingly, major differences in device performances were obtained with these three isostructural complexes comprising dibenzoylmethane (dbm), acetylacetonate (acac) or pyridine-2-carboxylic acid (pic) as the ancillary ligands. Best OLEDs were fabricated with the dibenzoylmethane-based iridium(III) complex. On the opposite, the picolate and the acetylacetonate-based complexes gave moderate performances. To complement this work, theoretical calculations were carried out on the three complexes. The newly synthesized complex i.e., Ir(tfpt)(pic) was also characterized by UV–vis absorption and luminescence spectroscopy as well as cyclic voltammetry.

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1. Introduction

Since the pioneering work of Forrest et al. phosphorescent organic light-emitting diodes (PHOLEDs) have been actively studied [1,2]. Contrarily to fluorescent organic light-emitting diodes that only utilize singlet excitons for light emission, PHOLEDs can harvest both singlet and triplet excitons to emit light, enabling to reach 100% internal quantum efficiency [3]. Among all phosphors that have been tested as triplet emitters for PHOLEDs, iridium complexes are undoubtedly the most widely studied [4,5]. Indeed, these complexes benefit from several appealing features such as high phosphorescent efficiencies at room temperature and relatively short luminescence lifetimes [6]. Emission color of these complexes can also be easily tuned from blue [7] to red [8] by modifying the ligands in the coordination sphere of the metal cation. As other figure of merit, neutral [7,8], cationic [9], anionic [10] complexes or even soft salts [11–13] composed of an anionic and a cationic iridium complex that can both participate to light emission were successfully used as emitters for light-emitting devices. However, neutral complexes are largely preferred over iridium salts, use of neutral complexes avoiding the presence of mobile counter-ions within the active layer. In the search for new

highly emissive triplet emitters, synthesis of new complexes bearing cyclometalated ligands other than the too much studied 2-phenylpyridine (ppy) is highly desired. In this regard, 1-methyl-5-phenyl-3-propyl-1*H*-1,2,4-triazole is a cyclometalated ligand that has only been scarcely investigated in the literature to design iridium(III) complexes [14,15]. However, this ligand possess several appealing features from the structure point of view. Notably, length of the alkyl chain can be easily tuned, enabling to prepare highly soluble complexes that could greatly help the fabrication of solution-processed devices in the future. The 1,2,4-triazole group can also be alkylated with a wide range of alkylating agents. To date, only five complexes were incorporated in OLEDs. As first examples, two wide bandgap homoleptic complexes comprising fluorinated tfpt ligands were tested as organometallic hosts for PHOLEDs. Interestingly, the most fluorinated complex furnished devices of improved performances compared to the reference devices comprising a conventional organic host e.g. 4,4-*N,N'*-dicarbazole-1,1'-biphenyl (CBP) [16]. Recently, two heteroleptic complexes i.e., Ir(tfpt)₂(dbm) and Ir(tfpt)₂(acac) (where tfpt stands for 5-(2,4,5-trifluorophenyl)-1-methyl-3-propyl-1*H*-1,2,4-triazole) were studied as dopants in multilayered devices [17]. While testing Ir(tfpt)₂(dbm) at the optimum dopant concentration of 8 wt %, devices reached a maximum luminance of 26150 cd/m² at 20 V, a maximum current efficiency of 19.3 cd/A and a maximum power efficiency of 8.1 lm/W. Prior to this work, lower performances were obtained with a dendrimers-like complex, reaching a maximum

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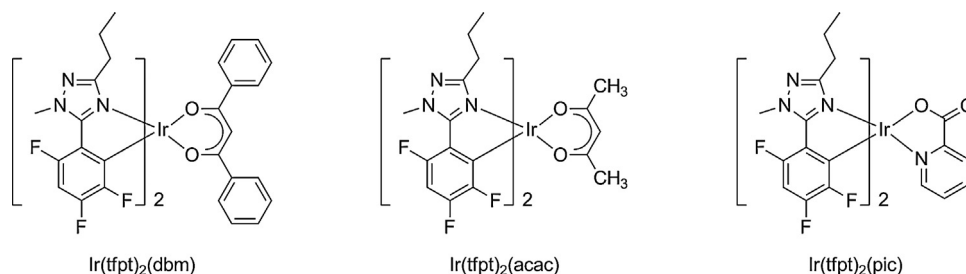


Fig. 1. Structure of complexes investigated in this study.

power and current efficiency of only 3.4 lm/W and 5.4 cd/A respectively [18]. In this work, three heteroleptic complexes differing by their ancillary ligands (dbm, acac, pic) were tested as dopants for PhOLEDs (see Fig. 1). Noticeably, $\text{Ir}(\text{tfpt})_2(\text{dbm})$ and $\text{Ir}(\text{tfpt})_2(\text{acac})$ that have already been studied in PhOLEDs were revisited in a new device configuration that furnished improved performances. Among the series, $\text{Ir}(\text{tfpt})_2(\text{dbm})$ proved to be the best emitter, followed by $\text{Ir}(\text{tfpt})_2(\text{pic})$ and $\text{Ir}(\text{tfpt})_2(\text{acac})$. To complement this work, theoretical calculations were also carried out to investigate the orbitals distribution of each dopant.

2. Experimental details

2.1. General information

^1H and ^{13}C NMR spectra were determined at room temperature in 5 mm o.d. tubes on a Bruker Avance 300 spectrometer equipped with a QNP probe head: ^1H (300 MHz) and ^{13}C (75 MHz). ^1H and ^{13}C NMR spectra were also determined at room temperature in 5 mm o.d. tubes on a Bruker Avance 400 spectrometer of the Spectropole: ^1H (400 MHz) and ^{13}C (100 MHz). The ^1H chemical shifts were referenced to the solvent peak: CDCl_3 (7.26 ppm), and the ^{13}C chemical shifts were referenced to the solvent peak: CDCl_3 (77.0 ppm). All starting materials and solvents were purchased from Aldrich or Lumtec and used as received commercially.

Cyclometalated iridium dimer $\text{Ir}_2(\text{tfpt})_4\mu\text{-Cl}_2$ (where tfpt stands for 5-(2,4,5-trifluorophenyl)-1-methyl-3-propyl-1H-1,2,4-triazole) was synthesized under inert atmosphere according to the Nonoyama route by refluxing $\text{IrCl}_3\cdot 3\text{H}_2\text{O}$ with 2–2.5 equiv. of cyclometalating ligand in a 3:1 mixture of 2-ethoxyethanol and water [19]. Absorption and emission spectra were recorded with a UV MC2 spectrophotometer from the SAFAS Monaco Society and a Photon Technology International spectrofluorimeter. Absolute fluorescence quantum yields in solution and in the solid state were measured by a Hamamatsu Photonics Quantaury QY at room temperature. Cyclic voltammetry (CV) was performed using an EG & G 273A driven by a PC with the M270 software at a scan rate of 100 mV/s, at room temperature in dichloromethane solution (10^{-3}M), with tetrabutylammonium hexafluorophosphate at 0.1 M concentration as the supporting electrolyte. All measurements were carried out in a nitrogen filled glovebox (O_2 and $\text{H}_2\text{O} < 0.1$ ppm). The working and the counter electrodes were a Pt disc and a Pt wire respectively, whereas Ag wire was used as a pseudo-reference electrode. Ferrocene was used as the internal standard material.

2.2. OLEDs fabrication and measurements

All materials used for the device fabrication were purchased from Lumtec with the best purity available and used as received.

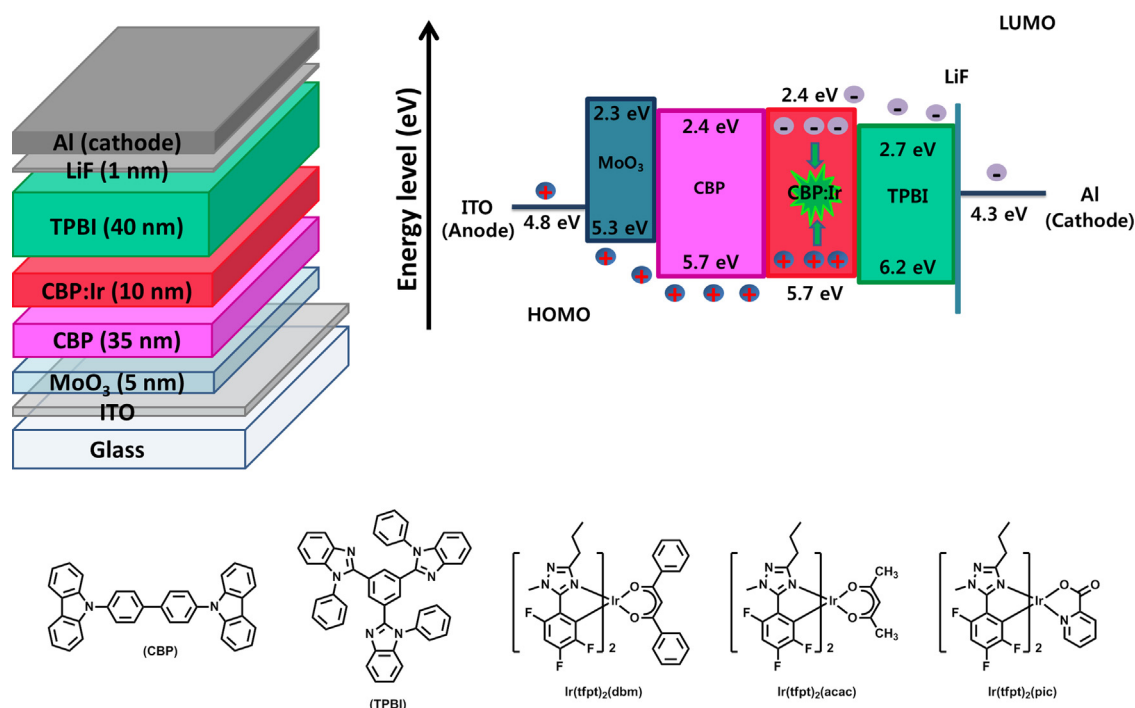


Fig. 2. Device structure, energy diagram and formulas of the different materials.

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