



Review

Synthesis, photophysical and electroluminescent properties of green organic light emitting devices based on novel iridium complexes containing benzimidazole ligands



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ABSTRACT

Synthesis, photophysical and electroluminescent analysis of cyclometalated iridium (III) complexes $\text{Ir}(\text{bpb})_2(\text{pic})$ are reported. The strongly allowed phosphorescence is the result of significant spin-orbit coupling of the Ir center. The lowest energy excited state is a mixture of metal to ligand charge transfer (MLCT) and $\pi-\pi^*$ states. Weak bands at longer wavelength were assigned to $^1\text{MLCT} \leftarrow S_0$ and $^3\text{MLCT} \leftarrow S_0$ transitions. These complexes were doped into emissive region of multilayer organic light emitting diode (OLED) by vapor-deposition emit green electroluminescence with similar current–voltage characteristics. These $\text{Ir}(\text{bpb})_2(\text{pic})$ doped OLEDs show appreciable efficiencies as compared with other iridium complex based OLEDs in the literature, which result from efficient trapping and radiative relaxation of singlet and triplet excitons formed in electroluminescent process. Devices with $\text{Ir}(\text{bpb})_2(\text{pic})$ shows the better performance in terms of brightness and power efficiency with brightness of 130301 cd/m^2 at 14.5 V and 26.2 lm/W at 5.5 V, respectively.

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Introduction

Third-row transition-metal complexes are well-known for their ability to achieve high-efficiency phosphorescence at room temperature [1,2] and has 100% internal quantum efficiency, because they can effectively harvest both singlet and triplet excitons [3]. As a result, Ru (II)-, Os (II)-, Ir (III)- and Pt (II)- based cyclometalated complexes were significantly developed which exhibited wide application in organic light-emitting devices (OLEDs) [4–7]. Among these, iridium (III) complexes display best electrophosphorescence with an external quantum efficiency as high as 27% in OLEDs and are considered to be a class of promising electrophosphorescent materials due to their non-planar configuration and short phosphorescent lifetime [8–14].

However, there is a crucial issue of phase separation between iridium (III) complexes and host materials that influence the doped device performance. This is overcome by increasing the bulkiness of the iridium (III) complexes which improved the dispersibility and high-efficiency emission results. Attempts were made to enhance the efficiencies of OLEDs by increasing the solubility and dispersibility of iridium (III) complexes [15–20]. Emission color of Ir (III) complexes is strongly governed by the nature of cyclometalating ligand and also by ancillary ligands [21–23]. The emission colors of Ir(dfppy)₂(LX) complexes are tuned from blue to red by changing ancillary ligands [24,25]. The non-radiative decay of Ir(ppz)₃ at ambient temperature are possible through higher thermally activated metal-to-ligand charge transfer (MLCT) states or MLCT to metal dd state conversion [26,27]. The picolinate complex Ir(ppz)₂(pic) shows relatively stronger green emission (526 nm) at room temperature whereas the emission colors change from blue (422 nm) to orange-red (587 nm) at low temperature (77 K) [28]. Despite elegant research on green phosphores [29–32], there are only few reports on room temperature green phosphores. The energy gap has been tuned by incorporating the substituents in the ligand to obtain the desired emission. The purpose of the present study is the molecular design of a highly efficient green-phosphorescent complexes viz., iridium (III) bis(1-benzyl-2-phenyl-1H-benzimidazolato-N,C^{2'})(picolinate), [Ir(bpb)₂(pic)] (**1**), iridium (III) bis(1-(4-fluorobenzyl)-2-(4-fluorophenyl)-1H-benzimidazolato-N,C^{2'})(picolinate) [Ir(fbfpb)₂(pic)] (**2**), iridium (III) bis(1-(4-methoxybenzyl)-2-p-tolyl-1H-benzimidazolato-N,C^{2'})(picolinate) [Ir(mbtb)₂(pic)] (**3**) and iridium (III) bis(1-(4-methoxybenzyl)-2-(4-methoxyphenyl)-1H-benzimidazolato-N,C^{2'})(picolinate) [Ir(mbmpb)₂(pic)] (**4**) suitable for green OLED devices. In addition to high phosphorescence efficiency, the complexes should possess high thermal stability for device fabrication and stable device performance. We particularly focused our attention on designing metal complexes that provide green emission from the MLCT excited state.

Experimental

Optical measurements and compositions analysis

The ultraviolet–visible (UV–vis) spectra of the phosphorescent iridium complexes were measured in an UV–vis spectrophotometer (Perkin Elmer Lambda 35) and corrected for background absorption due to solvent. Photoluminescence (PL) spectra were recorded on a (Perkin Elmer LS55) fluorescence spectrometer. NMR spectra were recorded on Bruker 400 MHz NMR spectrometer. The mass spectra of the samples were obtained using a Thermo Fischer LC-Mass spectrometer. Cyclic voltammetry (CV) analysis were performed by using CHI 630A potentiostat electrochemical analyzer. Measurements of oxidation and reduction were undertaken using 0.1 M tetra(n-butyl)ammonium–hexafluorophosphate

as the supporting electrolyte, at scan rate of 0.1 VS⁻¹. The potentials were measured against an Ag/Ag⁺ (0.01 M AgNO₃) reference electrode using ferrocene/ferrocenium (CP₂Fe/CP₂Fe⁺) as the internal standard. The onset potentials were determined from the intersection of two tangents drawn at the rising current and background current of the cyclic voltammogram. All calculations were performed using density functional theory (DFT) as implemented in the Gaussian-03 program using the Becke3-Lee-Yang-Parr (B3LYP) functional supplemented with the standard 6-31G (d, p) basis set [33].

Device fabrication

The EL devices based on the iridium (III) complexes were fabricated by vacuum deposition of the materials at 5×10^{-6} torr onto a clean glass precoated with a layer of indium tin oxide (ITO) as the substrate. The glass was cleaned by sonication successively in a detergent solution, acetone, methanol and deionized water before use. Organic layers were deposited onto the substrate at a rate of 0.1 nm s⁻¹. LiF and Alq₃ were thermally evaporated onto the surface of organic layer. The thickness of the organic materials and the cathode layers were controlled using a quartz crystal thickness monitor. A series of devices (**I**, **II**, **III** and **IV**) with the multilayer configuration ITO/NPB (30 nm)/iridium complex: CBP (7%) (30 nm)/BCP (10 nm)/Alq (40 nm)/Mg:Ag was fabricated. Measurements of current, voltage and light intensity of these devices (**I–IV**) were made simultaneously using a Keithley 2400 sourcemeter. The EL spectra and luminance of the devices were carried out in ambient atmosphere without further encapsulations.

General procedure for the synthesis of iridium complexes

A mixture of corresponding aldehyde (2 mmol), *o*-phenylenediamine (1 mmol) and ammonium acetate (2.5 mmol) has been refluxed at 80 °C in ethanol which yields the benzimidazole derivatives. The benzimidazole based cyclometalated iridium complexes have been synthesized via Nonoyama route [34a] (Scheme 1).

Iridium(III)bis(1-benzyl-2-phenyl-1H-benzimidazolato-N,C^{2'})(picolinate), [Ir(bpb)₂(pic)], (**1**)

Yield: 88%. ¹H NMR (400 MHz, CDCl₃): δ 5.72 (d, 1H, J = 8.0 Hz), 5.81–5.97 (m, 3H), 6.35 (d, 1H, J = 7.2 Hz), 6.32–6.73 (m, 3H), 6.78–6.88 (m, 2H), 7.16–7.54 (m, 16H), 7.86–7.95 (m, 2H), 8.14 (s, 1H), 8.25 (d, 1H, J = 7.6 Hz). ¹³C NMR (100 MHz, CDCl₃): δ 48.12, 48.37, 109.58, 110.29, 114.45, 117.68, 120.67, 121.39, 122.88, 123.51, 123.74, 124.62, 124.78, 124.86, 125.94, 126.01, 127.33, 127.68, 128.09, 128.02, 129.27, 129.33, 129.49, 129.83, 134.26, 134.36, 134.60, 135.06, 135.23, 135.28, 135.69, 137.28, 139.80, 149.17, 149.94, 153.57, 163.28, 164.68, 173.55. Anal. calcd. for C₄₆H₃₄IrN₅O₂: C, 62.71; H, 3.89; N, 7.95. Found: C, 62.67; H, 3.90; N, 7.98. MS: *m/z* 882.1, calcd. 881.23.

Iridium(III)bis(1-(4-fluorobenzyl)-2-(4-fluorophenyl)-1H-benzimidazolato-N,C^{2'})(picolinate), [Ir(fbfpb)₂(pic)], (**2**)

Yield: 90%. ¹H NMR (400 MHz, CDCl₃): δ 5.65 (d, 1H, J = 8.4 Hz), 5.83–5.92 (q, 5H), 6.23 (d, 1H, J = 10.4 Hz), 6.46–6.60 (m, 2H), 6.84–7.49 (m, 22H), 7.93 (d, 2H, J = 8.4 Hz), 8.09 (s, 1H), 8.28 (d, 1H, J = 7.2 Hz). ¹³C NMR (100 MHz, DMSO): δ 46.36, 46.59, 108.34, 111.58, 111.93, 113.21, 115.62, 115.68, 115.84, 115.90, 119.17, 123.77, 124.18, 124.27, 126.88, 128.04, 128.85, 130.94, 132.30, 135.11, 135.34, 138.52, 139.03, 149.08, 152.07, 152.82, 160.28, 161.65, 162.48, 162.68, 172.17. Anal. calcd. for C₄₆H₃₀F₄IrN₅O₂: C, 57.95; H, 3.18; N, 7.37. Found: C, 57.98; H, 3.17; N, 7.35. MS: *m/z* 954.1, calcd. 953.20.

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