

Synthesis and green electrophosphorescence of a novel cyclometalated iridium complex in polymer light-emitting diodes

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Received 28 June 2005; received in revised form 20 October 2006; accepted 23 October 2006

Available online 30 November 2006

Abstract

In this paper, we synthesized a new complex bis(dibenzo[f,h]quinolinato-*N,C*^{2'}) iridium(III) acetylacetonate ((*DBQ*)₂Ir(acac)) having a longer conjugate system than bis(2-phenylpyridinato-*N,C*^{2'}) iridium(III) acetylacetonate ((*PPY*)₂Ir(acac)). Interestingly, (*DBQ*)₂Ir(acac) emits at the same wavelength as (photoluminescence of 530 nm) (*PPY*)₂Ir(acac). A high-efficiency electrophosphorescent polymer light-emitting diodes was constructed by using (*DBQ*)₂Ir(acac) as the dopant, and a blend of poly(vinylcarbazole) (PVK) with 2-*tert*-butylphenyl-5-biphenyl-1,3,4-oxadiazol (PBD) as the host material. The electroluminescence efficiency of 9.5 cd/A is reported for the device doped with 3 wt% of (*DBQ*)₂Ir(acac). In this device, the emission from the PVK or PBD host was effectively inhibited with the using (*DBQ*)₂Ir(acac). Emission from the dopant molecules in such devices involve localization of the injected electron and hole on the metal-organic center. This can occur by a variety of mechanisms, including Förster and Dexter energy transfer from the host transport material to the dopant, and direct trapping of both electrons and holes on the metal-organic center.

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Keywords: Green emitter; PLEDs; Iridium

1. Introduction

Electroluminescence from light-emitting diodes made from small molecules figures in the history of flat panel display. Recently, highly efficient organic light-emitting diodes (OLEDs) using phosphorescent dyes such as (*PPY*)₂Ir(acac); bis(benzo[h]quinolinato-*N,C*^{2'}) iridium(III) acetylacetonate, (*Bzq*)₂Ir(acac) and their derivatives have been reported [1–4]. While (*PPY*)₂Ir(acac) exhibits green (530 nm) electroluminescence, (*Bzq*)₂Ir(acac) gives yellow (552 nm) electroluminescence. The two OLEDs have high external quantum efficiency ranging from 6% to 12.3%. The difference in the emission wavelength was related to the strength of the conjugate system; (*Bzq*)₂Ir(acac) has a stronger (longer) conjugate system than

(*PPY*)₂Ir(acac), and thus, emits at a longer wavelength. Based on this, the emission wavelength of a Ir(III) complex could be further shifted to even longer wavelength if the conjugate strength is further enhanced. (*DBQ*)₂Ir(acac) is structurally similar to but has a stronger conjugate system than (*PPY*)₂Ir(acac) and (*Bzq*)₂Ir(acac). In theory, it should emit in the orange-red region. In this work, (*DBQ*)₂Ir(acac) was synthesized and its photophysical properties were investigated in polymer lighting emitting diodes.

Recently, the photophysics of phenylpyrazolyl-based iridium complex Ir(*PPZ*)₃ has been the subject of extensive studies [1,5]. It has been shown that Ir(*PPZ*)₃ can be used as an electron/exciton blocking material (to prevent electron leakage to the hole-transporting layer) for constructing highly efficient single-dopant white OLED [5], and the *fac*-Ir(*PPZ*)₃ also exhibits intense luminescence at 77 K and room temperature via phosphorescence of a mixed ligand-centered-MLCT (LC-MLCT) triplet state

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[1,2]. In literature, however, it has been shown that $(PPY)_2Ir(acac)$ generally exhibits the similar photophysical properties as the $Ir(PPY)_3$ complex but the $(PPY)_2Ir(acac)$ is easier and less expensive to synthesized than $Ir(PPY)_3$ [4]. In this view, we synthesized in this work the bis(1-phenylpyrazolato- N,C^2') iridium(III) acetylacetonate, $(1-PPZ)_2Ir(acac)$, in place of $Ir(PPY)_3$ for OLED purpose. Furthermore, a methyl group was introduced to the 3' position of the phenylpyrazolyl fragment to produce, bis(3-methyl-1-phenylpyrazolato- N,C^2') iridium(III) acetylacetonate, $(3-MPPZ)_2Ir(acac)$ in an attempt to adjust the efficiency and color purity of the iridium complex-based EL devices.

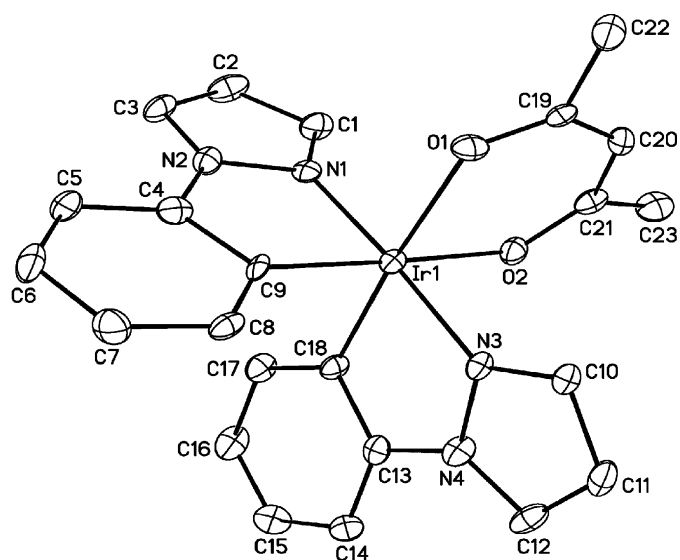


Fig. 1. A diagram showing the molecular structure of $(1-PPZ)_2Ir(acac)$.

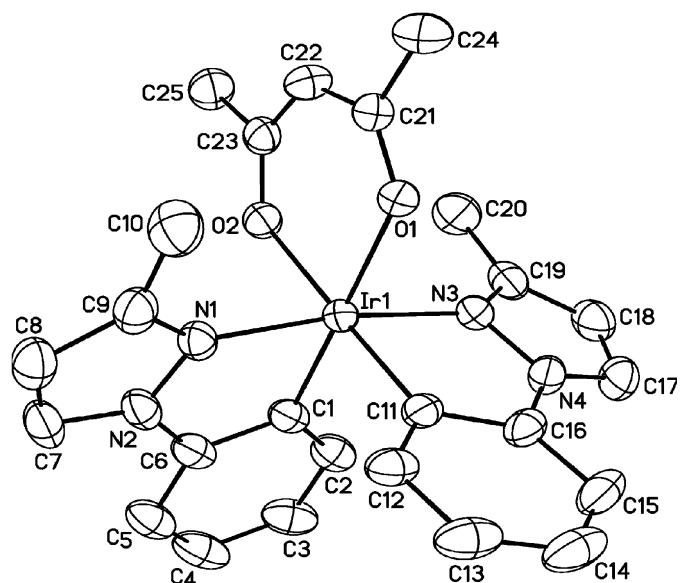


Fig. 2. A diagram showing the molecular structure of $(3-MPPZ)_2Ir(acac)$.

2. Experimental

All ligands were purchased from Aldrich and TCI, and were used without further purification. 1H -NMR and ^{13}C -NMR spectra were recorded on Bruker Avance-300 (300 MHz) or AMX-400 (400 MHz) spectrometer and the sample solution is CD_2Cl_2 ; chemical shifts are given in ppm and are reference to tetramethylsilane (TMS). The EI-Mass spectra were recorded on Bruker APEX II. UV-Vis spectra were recorded in CH_2Cl_2 solution on Agilent 8453 spectrometer, and the photoluminescence (PL) spectra were measured in CH_2Cl_2 solution with a HITACHI model F-2500 fluorescence spectrophotometer. HRMS spectra were obtained using a MAT-95XL resolution mass spectrometer. Elemental analyses have been carried out by using an Elementar Vario EL III analyzer.

3. Synthesis

3.1. Synthesis of $(DBQ)_2Ir(\mu-cl)_2Ir(DBQ)_2$ (as shown in Scheme 1) [6]

To a flask containing $IrCl_3 \cdot nH_2O$ (0.13 g, 0.44 mmole) and dibenzo[f,h]quinoline (DBQ) (0.25 g, 1.09 mmole) was added a 3:1 mixture of 2-ethoxyethanol and water. The mixture was refluxed at $110^\circ C$ for 24 h under nitrogen atmosphere and cooled to room temperature. After cooling, the product was isolated by filtration and washed with several portions of distilled water, *n*-hexane and ethylether. The solid was pumped to dry to give crude $(DBQ)_2Ir(\mu-Cl)_2Ir(DBQ)_2$ in 0.28 g (93%).

3.2. Synthesis of $(DBQ)_2Ir(acac)$

Crude $(DBQ)_2Ir(\mu-Cl)_2Ir(DBQ)_2$ (0.28 g, 0.21 mmole) was mixed with Na_2CO_3 (0.25 g, 2.31 mmole) in a flask. 2-ethoxyethanol (5.5 mL) and 2,4-pentanedione (0.11 g, 1.26 mmole) were added and the mixture was refluxed at $126^\circ C$ for 15 h. The product was cooled to room temperature then isolated by filtration and washed with distilled water, *n*-hexane and ether (77.4% crude product yield). The crude product was chromatographed using Benzene/EtOAc (6:1) to afford yellow orange powdery $(DBQ)_2Ir(acac)$. Anal. Calcd for $C_{39}H_{27}IrN_2O_2 \cdot n$ -hexane: C, 64.83; H, 4.92; N, 3.36. Found: C, 64.61; H, 5.22; N, 3.00. 1H -NMR (CD_2Cl_2 , 400 MHz) δ 8.9 (d, $J = 8.4$ Hz, 2H), 8.84 (dd, $J = 3.8, 1.2$ Hz, 2H), 8.73 (m, 2H), 8.69 (m, 2H), 7.92 (d, $J = 7.8$ Hz, 2H), 7.73 (m, 6H), 7.00 (td, $J = 7.8, 1.8$ Hz, 2H), 6.41 (dd, $J = 6.9, 1.7$ Hz, 2H), 5.28 (s, 1H), 1.78 (s, 6H). ^{13}C -NMR (CD_2Cl_2 , 100 MHz) δ : 175.1, 149.2, 137.6, 135.1, 130.9, 121.8, 121.7, 121.6, 121.1, 119.0, 118.6, 118.3, 117.3, 115.6, 114.0, 113.7, 111.6, 104.4, 90.6, 18.5. FABMS: m/z 649, $[M]^+$. HREIMS Calcd for $C_{39}H_{27}IrN_2O_2$: 748.1702. Found 748.1705.

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