

Highly efficient iridium(III) complexes with diphenylquinoline ligands for organic light-emitting diodes: Synthesis and effect of fluorinated substitutes on electrochemistry, photophysics and electroluminescence

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Received 4 May 2006; received in revised form 22 June 2006; accepted 7 July 2006

Available online 18 July 2006

Abstract

A series of novel cyclometalated iridium(III) complexes bearing 2,4-diphenylquinoline ligands with fluorinated substituent were prepared and characterized by elemental analysis, NMR and mass spectroscopy. The cyclic voltammetry, absorption, emission and electroluminescent properties of these complexes were systematically investigated. Electrochemical studies showed that the oxidation of the fluorinated complexes occurred at more positive potentials (in the range 0.57–0.69 V) than the unfluorinated complex **1** (0.42 V). In view of the energy level, the lowering of the LUMO by fluorination is significantly less than that of the HOMO. The weak and low energies absorption bands in the range of 300–600 nm are well resolved, likely associated with MLCT and $^3\pi-\pi^*$ transitions. These complexes show strong orange red emission both in the solution and solid state. The emission maxima of the fluorinated complexes showed blue shift by 9, 24 and 15 nm for **2**, **3** and **4**, respectively, with respect to the unfluorinated analogous **1**. Multilayered organic light-emitting diodes (OLEDs) were fabricated by using the complexes as dopant materials. Significantly higher performance and lower turn-on voltage were achieved using the fluorinated complexes as the emitter than that using the unfluorinated counterpart **1** under the same doping level. OLED devices using complexes **2** and **3** as the phosphorescent dopant at 3 wt% doping level exhibit very high performance. To complex **2**, the maximum luminance is 16410 cd/m² at a current density of 210 mA/cm², and the maximum luminance efficiency and power efficiency are 9.34 cd/A and 5.20 lm/W, respectively, with the emission of 605 nm. To complex **3**, those data are 16797 cd/m² at a current density of 211 mA/cm², 11.12 cd/A and 4.97 lm/W, respectively, with the emission of 593 nm.

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Keywords: Iridium complexes; Phenylquinoline; Fluorination; Electrochemistry; Photophysics; Electroluminescence

1. Introduction

Recently, the iridium complexes as phosphorescent emitter in organic light-emitting diodes (OLEDs) have attracted much attention since the realization of a high efficiency OLED device based on the complex *fac*-tris(2-phenylpyridine)iridium [Ir(ppy)₃] [1]. The frequency of emission of the iridium complexes can usually be tuned by modification or variation of cyclometalated ligands of 2-phenylpyridine or its analogical ligands, such as benzoisoquinolines [2], 2-phenylbenzothiazole [3], benzoimidazole [4], etc. Several groups have studied the mechanism of the OLEDs based on phosphorescent heavy metal complexes [5]. It has been demonstrated that the Förster energy transfer plays minor role in achieving high efficiency in

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these devices, instead that direct charge trapping plays dominant role in electroluminescence. Therefore, in order to obtain high efficiency OLED devices, it would be desirable to design the phosphorescent complexes as energy acceptors, more importantly, as traps for holes or/and electrons.

Quinoline-based compounds have received considerable attention in optoelectronic materials due to their high electron affinities. For example, aluminum tris(8-hydroxyquinolate) (AlQ₃) has been usually applied as electron transporting, electron-emitting and host materials in doped OLED systems. Most recently, phenylquinoline or phenylisoquinoline based iridium complexes have been proven to be good red emitters [2,6]. On the other hand, iridium complexes carrying fluorinated phenylpyridyl ligands have shown several benefits, such as enhancing the photoluminescence efficiency and improving the sublimation [7].

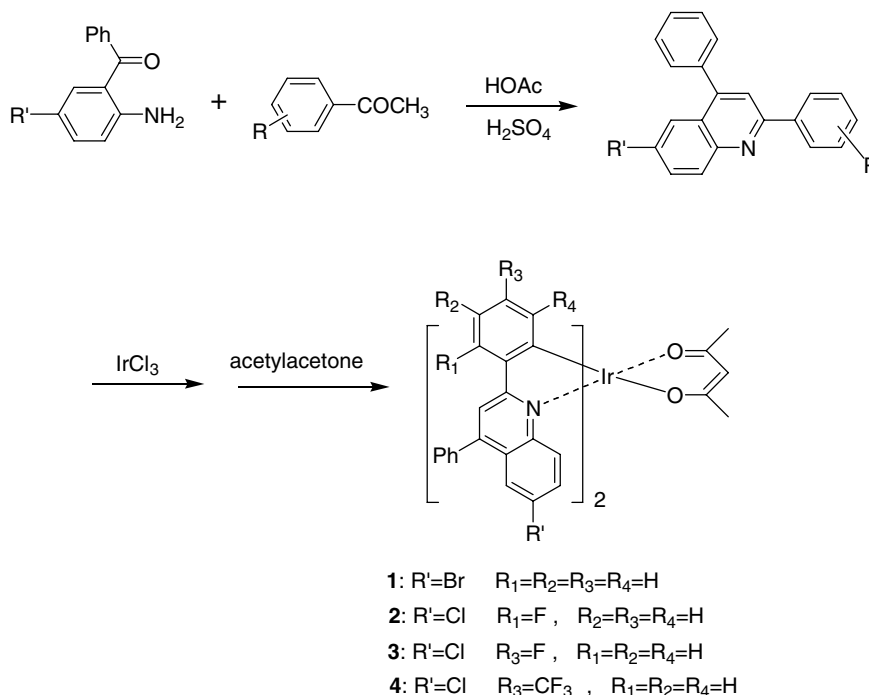
In this paper, we report a series of novel iridium complexes bearing 2,4-diphenylquinoline ligands with strong electron-withdrawing fluorinated substituent, as well as the unfluorinated analogue for comparison. We expect that the phenylquinoline ligands modified by fluorinated substituent could improve the electron transport ability of the complexes, consequently to facilitate the charge trapping across the bulk for high performance OLEDs [8]. Meanwhile, it will be significant to study the effect of fluorinated substituent on the electrochemistry, photophysics and electroluminescent performance of iridium complexes in order to understand the relationship between the structures and properties. In addition, the bulky phenyl group in the position 4 of quinoline ring may prevent the crystallization and

suppress the aggregation-forming tendency, and the chloride or bromide in the position 6 of quinoline ring may increase luminescent efficiency by heavy-atom effect.

2. Results and discussion

2.1. Synthesis and characterization

Phenylquinoline-based organic ligands were conveniently prepared from 5-chloro-2-aminobenzophenone (or 5-bromo-2-aminobenzophenone) and corresponding acetophenone derivatives through Friedländer reaction in moderate yields (Scheme 1) [9]. The Ir(III) μ -chloro-bridged dimers were synthesized by the reaction of iridium trichloride hydrate with ligands according to a conventional procedure [10]. Then the diiridium complexes were converted to mononuclear iridium complexes by replacing the two bridging chlorides with bidentate monoanionic acetylacetonate ligand in 50–70% yields. Elemental analysis of each of the four complexes is consistent with the expected formulation of their structures. The mass spectra give corresponding molecular ion peaks at 1010 for **1**, 956 for **2** and **3**, and 1056 for **4**. In the ¹H NMR, the acetylacetonate CH protons appear in the region 4.69–4.79 ppm as a sharp singlet, obviously lower than the corresponding signals usually appeared at >5.1 ppm in the other iridium complexes with arylpyridine ligands. This is presumably due to the weaker ligand field strength of quinoline that inhibits effective back-donation from metal center to ligand, consequently, metal center has larger electron density which more shields the acetylacetonate CH protons [6].



Scheme 1. Synthesis of the iridium complexes.

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