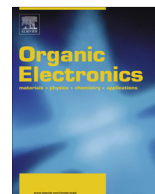




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New deep-red heteroleptic iridium complex with 3-hexylthiophene for solution-processed organic light-emitting diodes emitting saturated red and high CRI white colors

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ABSTRACT

The exploitation of soluble and efficient deep-red phosphorescent emitters is of paramount importance for solution-processed organic light-emitting diodes (OLEDs) applied in both high-quality RGB displays and high color-rendering-index (CRI) solid-state lighting source. In this work, a new deep-red heteroleptic iridium(III) complex, i.e. bis[2,5-di(4-hexylthiophen-2-yl)pyridine][acetylacetonate]iridium(III) [Ir(ht-5ht-py)₂(acac)], has been synthesized and successfully used to fabricate solution-processed saturated red and white organic light-emitting diodes (WOLEDs). The long alkyl side-chains of Ir(ht-5ht-py)₂(acac) render its excellent solubility in common organic solvents and good compatibility with common host materials. The solution-processed red OLED based on Ir(ht-5ht-py)₂(acac) exhibited a decent external quantum efficiency of 8.2% and a power efficiency of 6.5 lm/W, with satisfactory Commission International de L'Eclairage (CIE) coordinates of (0.68, 0.31) for saturated red emission. Furthermore, the prepared multiple-phosphors-doped WOLED with Ir(ht-5ht-py)₂(acac) as the red emitter showed an excellent high color rendering index (CRI) value of 89 as well as low color-correlated temperature (CCT) of 2331 K, which can meet the call for physiologically-friendly indoor illumination.

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1. Introduction

Solution-processed phosphorescent organic light-emitting diodes (OLEDs) have drawn great attention in the past decades since they hold great potential in large-area and cost-effective manufacturing of flat panel displays and solid-state lighting sources. The efficient phosphorescent emitters that can harvest both singlet and triplet excitons for radiative decay are of paramount importance for

achieving high device performance [1–4]. Efficient and solution-processible deep red phosphors and devices are indispensable in various kinds of organic electroluminescent (EL) devices such as red–green–blue (RGB) full color displays and white lighting devices [5]. It not only functions as a primary color for high color quality RGB displays, but also plays an important role in determining the light-emitting efficiency and color quality of white light-emitting devices (WOLEDs) [6–7]. Moreover, the widely used physiologically-friendly “candle-like” white lighting devices also require efficient deep red phosphors to achieve high color rendering index (CRI) and low color correlated temperature (CCT) merits [5]. Although great progress has been achieved

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in solution-processed red OLEDs by using novel host materials in combination with typical light-red dopants [9–11], deep-red phosphors and devices with CIE- $x \geq 0.67$ are still scarce up to now [12,13].

Several saturated red phosphors have been developed previously to improve the light-emitting efficiencies of the solution-processed red OLEDs. For example, Park et al. synthesized an efficient red phosphor [2-(9,9-dihydro-9H-fluoren-2-yl)-4-phenylquinoline]₂ iridium(III) picolinic acid *N*-oxide [(FPQ)₂Ir(pic-N-O)] and successfully applied it in solution-processed red OLEDs, achieving a luminous efficiency (LE) of 9.9 cd/A, a power efficiency (PE) of 3.9 lm/W and an external quantum efficiency (EQE) of 8.9%, with the CIE coordinates of (0.660, 0.338) [14]. We have reported an efficient saturated deep-red iridium dendrimer containing arylamine units as periphery dendron to realize superior efficiency/color purity trade-offs [10]. The resultant device exhibited a high EQE of 11.65% and a PE of 3.65 lm/W with a CIE coordinate of (0.70, 0.30). In contrast to the dendritic route to develop solution-processible red phosphors [15,16], Chao et al. proposed to modify red tris(1-phenylisoquinoline) iridium [Ir(piq)₃] with long side chain to improve its solubility [17], thus facilitating the miscibility with common poly(vinylcarbazole) (PVK) host and leading to a distinct efficiency increase from 0.74 cd/A to ca. 6 cd/A. In spite of these improvements, highly-soluble efficient deep-red phosphors remain scarce and limit the development of high color quality solution-processed red OLEDs and WOLEDs [18–20]. As an example, the CRI values of these WOLEDs were typically less than 80 and thus did not meet the practical requirement for white color lighting.

Here, we synthesized a new solution-processible red heteroleptic iridium(III) complex, i.e. bis[2,5-di(4-hexylthiophen-2-yl)pyridine][acetylacetonate]iridium(III) [Ir(ht-5ht-py)₂(acac)], which showed pure red photoluminescence (PL) emission with peak located at 628 nm, corresponding to the CIE coordinates of (0.68, 0.31). The red phosphor [Ir(ht-5ht-py)₂(acac)] exhibits good solubility in common organic solvents and excellent miscibility with host in solution process. The corresponding solution-processed red OLED showed a promising EQE of 8% and a PE of 6.5 lm/W. Furthermore, by combining it with a blue phosphorescent iridium(III) [bis(4,6-difluorophenyl)-pyridinato-N,C²]-picolinate [Irpic] [21], green phosphorescent G0 [22] and orange phosphorescent dopant containing 5-trifluoromethyl-2-(9,9-diethylfluoren-2-yl)pyridine ligand [Ir(Flpy-CF₃)₃] [20] to prepare four-color white emissive layer, the resulting WOLEDs show a high CRI of 89 and low CCT of 2331 K, making it a physiologically-friendly white lighting source [8]. This is among a few reports that solution-processed WOLEDs achieve such ideal warm white light emission.

2. Experimental

2.1. General information

All chemicals and reagents were purchased from Aldrich Chemicals Company. The solvents were carefully dried and distilled with appropriate drying agents prior to use. Commercially available reagents were used without

further purification unless otherwise stated. The ¹H and ¹³C NMR spectra were recorded with Bruker Advanced 400 MHz NMR spectrometer. Thermal gravimetric analysis (TGA) was performed on Perkin-Elmer-TGA 7 thermal gravimetric analyzer under nitrogen flow at a heating rate of 10 °C/min. MALDI-TOF-TOF was carried out using Bruker autoflex III smart beam mass spectrometer.

2.2. Synthesis of ligand

2,5-Dibromopyridine (2.0 g, 7.55 mmol), 3-hexylthiophene boronic acid (4.0 g, 18.9 mmol) and Pd(PPh₃)₄ (262.0 mg) were added into a mixture of THF (30 mL) and 2 M Na₂CO₃ (8 mL) under N₂ atmosphere. The reaction mixture was heated to 90 °C for 48 h with stirring. Then the reaction mixture was cooled down to room temperature and extracted with ethyl acetate (EA). The combined organic phase was washed with water. The organic phase was separated and dried over MgSO₄. The solvent was removed under reduced pressure and the residue was purified by column chromatography eluting with CH₂Cl₂/hexane. The product was obtained as a white crystalline solid. ¹H NMR (400 MHz, CDCl₃): δ (ppm) 8.80 (d, *J* = 1.9 Hz, 1H, Ar), 7.84 (d, *J* = 6.8 Hz, 1H, Ar), 7.61 (d, *J* = 8.3 Hz, 1H, Ar), 7.51 (d, *J* = 4.5 Hz, 1H, Ar), 7.20 (d, *J* = 1.0 Hz, 1H, Ar), 6.99 (d, *J* = 11.8 Hz, 1H, Ar), 6.94 (s, 1H, Ar), 2.63 (t, *J* = 7.7 Hz, 4H, hexyl), 1.72–1.59 (m, 4H, hexyl), 1.42–1.23 (m, 12H, hexyl), 0.90 (dd, *J* = 7.0, 5.6 Hz, 6H, hexyl). ¹³C NMR (101 MHz, CDCl₃): δ (ppm) 151.31, 146.39, 144.70, 144.47, 143.93, 140.06, 133.26, 128.69, 125.96, 125.19, 122.48, 120.42, 118.55 (Ar), 31.70, 30.65, 30.58, 30.11, 29.01, 22.64, 14.12 (hexyl).

2.3. Preparation of Ir(ht-5ht-py)₂(acac)

The phosphorescent iridium complexes were prepared according to the well-established two-step strategy from the cyclometalation of IrCl₃·3H₂O with the corresponding organic ligand to form, initially, the μ-chloro-bridged dimer, followed by coordination of the acetylacetonate (acac) anion in the presence of Na₂CO₃ [23]. The reaction mixture was extracted with CH₂Cl₂. The combined organic phase was washed with water. The organic phase was separated and dried over MgSO₄. The product was purified by silica gel column chromatography with CH₂Cl₂/hexane as an eluent and a dark red solid was obtained. ¹H NMR (400 MHz, CDCl₃): δ (ppm) 8.49 (d, *J* = 1.8 Hz, 2H, Ar), 7.68 (dd, *J* = 8.5, 2.1 Hz, 2H, Ar), 7.36 (d, *J* = 8.4 Hz, 2H, Ar), 7.04 (t, *J* = 5.4 Hz, 2H, Ar), 6.81 (s, 2H, Ar), 6.69 (s, 2H, Ar), 5.21 (d, *J* = 4.1 Hz, 1H, acac), 2.52 (dd, *J* = 15.2, 7.4 Hz, 4H, alkyl), 1.85–1.66 (m, 9H, alkyl), 1.66–1.44 (m, 12H, alkyl), 1.34–1.09 (m, 28H, alkyl), 1.09–0.93 (m, 11H, alkyl), 0.93–0.72 (m, 19H, alkyl), 0.72–0.60 (m, 8H, alkyl). ¹³C NMR (101 MHz, CDCl₃): δ (ppm) 183.33 (acac), 163.07, 150.44, 146.59, 144.04, 143.56, 138.46, 135.54, 132.75, 124.08, 123.4, 121.51, 118.85, 115.52, 99.86 (Ar), 30.64, 30.48, 29.52, 29.36, 29.09, 28.68, 28.41, 27.95, 27.51, 21.66, 21.56, 13.07, 13.01 (alkyl + acac). Calcd for C₅₅H₇₇IrN₂O₂S₄: C, 59.37; H, 6.43; N, 2.52, found: C, 59.33; H, 6.92; N, 2.52. MALDI-TOF-TOF-MS: *m/z* found 1112.40, calcd 1112.66.

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