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A yellow-emitting iridium complex for use in phosphorescent multiple-emissive-layer white organic light-emitting diodes with high color quality and efficiency ☆

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Abstract

A cyclometalated iridium(III) complex containing 2-(9,9-diethylfluoren-2-yl)pyridine [**Ir(Flpy)$_3$**] was prepared and used in the fabrication of both yellow and white organic light-emitting diodes (OLEDs). A hole-blocking material has been used as a hole barrier layer in-between different emission layers, helping the formation of the hole limitation region. With the proper position of a hole barrier layer and the construction of a four-emission-layer structure involving the use of [**Ir(Flpy)$_3$**], the resulting WOLED shows sound device performance as well as very stable color even at high luminances. Such WOLEDs have been demonstrated to reveal superior white light color stability/efficiency trade-off optimization. The Commission Internationale de L'Eclairage (CIE) coordinate differences Δx and Δy are confined to ± 0.015 when the luminance increases from 13 to 14806 cd/m^2. The color rendering index (CRI) of the device is also very good, which varies only from 86 to 87 by changing from the normal direction to 80° off-normal at 12 V. The peak electrophosphorescence efficiency can reach as high as 24.6 cd/A at 168 cd/m^{2}and it can still be kept at 17.2 cd/A at 10834 cd/m^2. Such outstanding performance renders this all-phosphor WOLED very attractive as a white light source for illumination applications, which typically demand high efficiency, high CRI, and stable color in high brightness work conditions.

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1. Introduction

While the full-color organic light-emitting diodes (OLEDs) have gained tremendous increase in performance over the last decade [1], white OLEDs (WOLEDs) are currently of a growing interest to the lighting research community [2]. Non-pixelated, large area WOLEDs [3] find promising applications as backlights for the liquid crystal display technology and as light sources in the illumination system owing to their potentials for the superior light emission efficiency over traditional incandescent sources in addition to the low-cost and high-throughput manufacturability [4]. However, lighting applications are largely dictated by the need for the WOLED color to be at a particular color point on the blackbody locus as well as the requirement for provision of an adequate color rendering capability for objects viewed in the white light [5].

The precise control of dopants, the demand for an ultra-uniform mixture of colors and the ease of fabrica-

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☆ In memory of my mentor, Prof. F. Albert Cotton, for his remarkable contribution to inorganic and organometallic chemistry throughout his life.

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tion steps are all important factors for the WOLEDs to achieve an extremely little color shift with increasing injection current. The fabrication of WOLEDs is relatively more complicated than that of other monochromatic color devices and the color stability is difficult to be maintained at will since an accurate control of at least two dopant concentrations is required. Over the years, a number of device structural concepts have been employed to generate highly efficient white light electrophosphorescence. Among these, one of the most common approaches is to use three separate emitters, each emitting one primary color from red (R), green (G) and blue (B) [6,7]. While the fused organic solid solution method [8] generally requires a careful control over several dopants, it can reduce the fabrication complexity. Tandem (or stacked) WOLEDs consisting of multiple electroluminescent (EL) elements connected in series can also lead to a significant improvement in efficiency [9]. Recently, a novel concept that exploits a blue fluorescent molecule in exchange for a phosphorescent blue dopant furnishes more stable WOLED devices than those mixed with the blue triplet emitters [10]. Keeping a high color stability with a wide driving range and maintaining the EL efficiency at high luminances are still of prominent importance to overcome the hurdle faced nowadays in using WOLEDs as illumination sources and represent one of the major targets in WOLED research for the eventual lighting applications.

In this work, we have synthesized a new yellow (Y) phosphor $[Ir(L)_3]$ (HL = 2-(9,9-diethylfluoren-2-yl)pyridine) [**Ir**(**Flpy**)**3**] and demonstrated the exploitation of [**Ir(Flpy)**$_3$] in the fabrication of some WOLEDs with excellent performance in terms of color stability and efficiency. The device consists of a four-emissive-layer architecture (made of B, G, Y and R colors) sandwiched by some charge-injection and -blocking layers without the need for the more sophisticated stacked design [9]. The excitons are effectively confined by the properly interposed hole-blocking material. Saliently, the CIE color coordinate differences can be confined to within ±0.015 when the luminance intensity increases and the CRI value is high and remains almost invariant from the normal to the 80° off-normal direction. The forward emission EL efficiency is as high as 24.6 cd/A at 168 cd/m^{2}and can impressively be kept at ~70% of the peak value (ca. 17.2 cd/A) at a high brightness of 10834 cd/m^2.

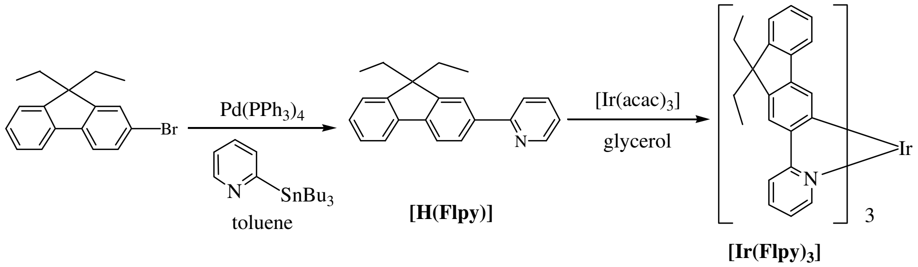
2. Results and discussion

2.1. Synthesis, chemical and photophysical characterization of [Ir(Flpy)$_3$]

Scheme 1 shows the synthetic protocol for the homoleptic iridium(III) complex [**Ir(Flpy)**$_3$]. The key compound in our studies is the cyclometalating ligand [**H(Flpy)**], obtained from the Stille coupling of 2-bromo-9,9-diethylfluorene [11] with 2-(tributylstannyl)pyridine in the presence of $[Pd(PPh_3)_4]$. The Ir complex was obtained by direct thermal reaction of $Ir(acac)_3$ with [**H(Flpy)**] in refluxing glycerol [12]. Purification of the mixture by silica chromatography furnished [**Ir(Flpy)**$_3$] as an air-stable orange powder in high purity. It was fully characterized by NMR spectroscopy and FAB-MS spectrometry. The first-order ^{1}H NMR spectrum of [**Ir(Flpy)**$_3$] is consistent with a facial geometry around the Ir center, which indicates that the number of coupled spins is equal to that of protons on one ligand because the three cyclometalating ligands are magnetically equivalent due to the inherent C_3 symmetry of the complex. The parent ion peak at $m/z = 1087$ amu in the FAB mass spectrum confirms its identity.

[**Ir(Flpy)**$_3$] is a highly amorphous and morphologically stable solid and has a high glass transition temperature (T_g) of 118 °C as revealed by differential scanning calorimetry (DSC). It is very thermally stable with onset decomposition temperature determined by thermogravimetric analysis (TGA) to be 441 °C. Also, it can be readily sublimed under vacuum before thermal decomposition was reached.

Photophysical examination of [**Ir(Flpy)**$_3$] reveals that it is characterized by strong absorption bands at $\lambda_{max} = 321$, 336 and 405 nm (Fig. 1). The intense absorptions ($\varepsilon \sim 4.7$–6.0×10^4) at $\lambda_{max} = 321$, 336 nm for [**Ir(Flpy)**$_3$] resemble those for the free ligand [**H(Flpy)**] ($\lambda_{max} = 305$ and 319 nm) and thus can be assigned to the spin-allowed ligand-centered $^1\pi$–π^* transitions. These $^1\pi$–π^* absorption bands below 400 nm are also accompanied by weaker, lower energy features peaking at 405 nm in the visible regime that tail toward 520 nm and they should correspond to an admixture of ^{1}MLCT, ^{3}MLCT and $^3\pi$–π excited states. The strong spin–orbital coupling induced by the heavy-atom effect of Ir center between the singlet and triplet manifolds can give rise to the ^{3}MLCT and $^3\pi$–π bands [13]. [**Ir(Flpy)**$_3$] emits a strong yellow phosphorescence



Scheme 1. The synthetic route to the yellow-emitting complex [**Ir(Flpy)**$_3$].

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