



A wet and dry processable phosphorescent green dye based organic light-emitting diodes



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ABSTRACT

Electroluminescence-efficiency green emitter with both wet- and dry-process feasibility is highly desirable to realize cost-effective large roll-to-roll manufacturing and high performance products. We demonstrate here a wet- and dry-process feasible green bis[5-methyl-8-trifluoromethyl-5H-benzoc(1,5)naphthyridin-6-one]iridium(picolate) containing organic light-emitting diode device with an efficacy roll-up character between 100 and 1000 cd/m². The newly synthesized iridium complex exhibits a relatively short excited-state lifetime (0.39 μs) and a high quantum yield of 74%, both which warrant this complex to be a highly electroluminescence active candidate. The wet processed device using a 3,5-di(9H-carbazol-9-yl) tetraphenylsilane host, for example, shows a 52 lm/W with an 18% external quantum efficiency (EQE) at 100 cd/m², which increases to 61 lm/W with a 23% EQE at 1000 cd/m². For the dry-processed device using a 4,4-bis(carbazol-9-yl)biphenyl host, it is 59 lm/W with a 25% EQE at 1000 cd/m².

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1. Introduction

Organic light-emitting diode (OLED) possesses high potential in high quality flat panel display and solid-state lighting applications [1–3]. Although numerous passive matrix and a few active matrix OLED devices have already in the market and their efficiency is sound, further improving the device efficiency is still very crucial for prolonging the device lifetime [4]. Especially, high efficiency green emission is always required for OLED to be commercially acceptable for illumination.

Having an electroluminescence (EL) efficient green emitter, one major component of the 3- or more-band composing white light sources, is highly critical to achieve high efficacy white light since luminous sensitivity of human eyes is the highest with green light and peaking at 555 nm, while becomes decreasingly sensitive drastically as approaching the deep-red or deep-blue region [5].

Having a high efficiency green emission would consequently be able to generate a high efficiency white light. It would be more ideal if any EL-effective green emitters possess both wet- and dry-process feasibility in order to realize, respectively, cost-effective large area size roll-to-roll manufacturing and high performance products.

Over the past years, phosphorescent materials have drawn increasing attention because of their ability to fully utilize both singlet and triplet excitons, achieving nearly 100% internal quantum efficiency [6–9], and based on which many record breaking efficient OLEDs have hence been fabricated [10–19]. For long wavelength emission devices, for examples, Chi's group reported an Osmium based red device with a 26 lm/W at 1000 cd/m² [10]. Jou's group reported an iridium based orange-red device with a 32 lm/W [11]. For green devices, Kido's group reported a 107 lm/W by using a dry-processed iridium based green emitter [12], Jou's group reported a 69 lm/W by using a different but wet-process feasible iridium based green emitter [13]. Helander's group reported a 79 lm/W by using chlorinated indium tin oxide electrodes, and efficacy increase to 155 lm/W by using out-coupling [15]. For blue devices, Kido's group reported a 46 lm/W by using an iridium based

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blue emitter [16]. By using the same blue emitter, Jou's group reported a 25 lm/W blue device with a solution-process feasible molecular host and enhanced by incorporating a polymeric nanodot in the hole injection layer [17]. Leo's group reported a 90 lm/W white OLED using iridium based red, green, and blue emitters coupling with the use of out-coupling [18]. Jou's group reported a 25 lm/W white OLED using a solution-processed emissive layer composing iridium based orange-red, blue-green, and blue emitters [19].

It is noteworthy that most of the dry-processed OLEDs show better device efficiency than the wet-processed counterparts. However, in some cases, the wet-processed devices show better device efficiency than their dry-processed counterparts [13,14,20], especially for those employing high molecular weight molecular hosts and guests. It would be ideal if an OLED material can be fabricated with high device efficiency by using either dry- or wet-process.

In this report, we demonstrate a both wet- and dry-process feasible green dye. By using the green dye, OLEDs with device efficiency at high luminance along with an extraordinary efficacy roll-up phenomenon were fabricated. Via wet-process, the resulting device exhibited an efficacy increasing from 52 to 61 lm/W, with the corresponding external quantum efficiency (EQE) increasing from 18% to 23%, as the luminance from 100 to 1000 cd/m², which approaches to the best device performance reported by Jou's group with power efficiency of 69 lm/W and EQE of 26% at 1000 cd/m² [13]. For the dry-processed counterpart, its efficacy remained at 59 lm/W with the EQE increasing from 21% to 25%. It is noteworthy that by using the best known ultra-high efficiency green dye, tris(2-phenylpyridine) iridium(III), Ir(ppy)₃ [12,21–26], the resultant devices show an efficacy of 20 lm/W and EQE of 8% at 100 cd/m², or 12 lm/W and 6% EQE at 1000 cd/m² by using wet-process, while 39 lm/W and 6% EQE at 100 cd/m², or 25 lm/W and 5% EQE at 1000 cd/m² by using dry-process, based on the same device structure. These results indicate this green dye presented herein to possess potential to yield an even higher efficiency if better hole and electron transporting materials, as reported in the literature, could be applied.

2. Green emitter synthesis

Scheme 1 shows the synthesis of the pure green light-emitting bis[5-methyl-8-trifluoromethyl-5H-benzo(c)(1,5)naphthyridin-6-one] iridium (picolinate) (3-CF₃BNO). Iridiumtrichloride hydrate was first reacted with 5-methyl-8-trifluoromethyl-5H-benzo(c)[1,5]naphthyridin-6-one to give the corresponding cyclometalated Ir(III)-μ-chloro-bridged dimers. Further reaction of the dimers with picolinic acid and sodium carbonate formed the desired iridium complex, 3-CF₃BNO.

3. Green emitter characteristics

Table 1 lists the physical characteristics of the green light-emitting guest, 3-CF₃BNO, comparing against those of the comparing counterpart, Ir(ppy)₃. Also included are the physical characteristics of a previously reported blue-green light-emitting

Table 1

Physical characteristics of the novel green light-emitting dopant, 3-CF₃BNO, compared with those of the two counterparts, i.e. the blue-green CF₃BNO and green Ir(ppy)₃ emitters. Comparing with the most known extremely high efficiency green Ir(ppy)₃ emitter, the green 3-CF₃BNO emitter exhibits a relatively short excited-state lifetime and a high quantum yield. It also possesses a decomposition temperature much higher than its isomeric counterpart, 2-CF₃BNO, indicating its feasibility in dry-process using thermal evaporation.

Emitter	Emission	τ (μ s)	Φ_{em} (%)	MW (g/mol)	T_d ($^{\circ}$ C)
3-CF ₃ BNO	Green	0.39	72	869	385
2-CF ₃ BNO	Blue-green	0.30	95	869	290
Ir(ppy) ₃	Green	1.15	51	655	395

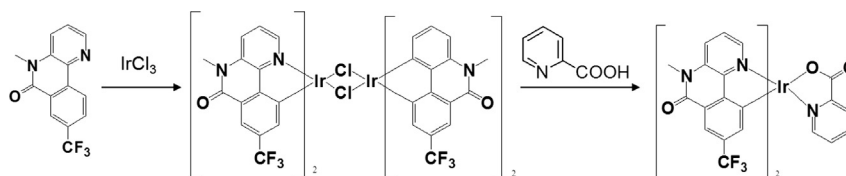
guest, bis[5-methyl-7-trifluoromethyl-5H-benzo(c)(1,5)naphthyridin-6-one]iridium(picolinate) (2-CF₃BNO) [14]. The excited-state lifetime for 3-CF₃BNO is 0.39 μ s, much shorter than the 1.15 μ s for Ir(ppy)₃. Similar to that, 0.30 μ s, observed in 2-CF₃BNO, the much shorter excited-state lifetime shown in 3-CF₃BNO should presumably favor the fabrication of a device with an efficiency higher than the Ir(ppy)₃ counterpart [14,27].

The quantum yield for 3-CF₃BNO in dimethoxyethane solution is 72%. Although lower than the 95% for 2-CF₃BNO, it still is much higher than the 51% for Ir(ppy)₃. The higher quantum yield of 3-CF₃BNO again indicates that its resulting device could be more efficient than the Ir(ppy)₃ counterpart [14,28].

According to the quantum chemical calculations using the density functional theory, the HOMO of BNO molecule is localized more at the vicinity of the 2-substitutional site, while the LUMO is localized more at the 3-substitutional site, as illustrated in parts (a) and (b) of Fig. 1. Overall speaking, the molecular orbital of the BNO molecule would be more stabilized upon introducing the electron-withdrawing CF₃ group at either the 2- or 3-substitutional site. For 2-substituted isomer, the resulting HOMO would be lowered more significantly than the LUMO does hence the band gap increases. As a result, when the CF₃ moiety is introduced at the 2-substitutional site, the emitter shows a much higher band gap and gives a bluish green emission, as comparing with the green 3-CF₃BNO emitter. In contrast, by introducing the same CF₃ moiety at the 3-substitutional site, the LUMO would be lowered more significantly than the HOMO does, which in turn leads to a smaller band gap. These explain why the 3-CF₃BNO isomer emits green light, while the 2-CF₃BNO blue-green. As shown in Fig. 1, the HOMO of such a BNO series is found to appear mainly at position 2 and the LUMO is localized at position 3. As a result, the HOMO and LUMO levels would be significantly affected as the substitute group, -CF₃, is changed from position 2 to 3, which would in turn affect the quantum chemical properties and likewise the corresponding quantum yield.

The 3-CF₃BNO has the same molecular weight as 2-CF₃BNO, which is 869 g/mol, much higher than the 655 g/mol for Ir(ppy)₃. The comparatively high molecular weight should favor the formation of an emissive layer with good film integrity in solution process [20].

Importantly, the 3-CF₃BNO exhibits a decomposition temperature of 385 $^{\circ}$ C, which is much higher than the 290 $^{\circ}$ C of 2-CF₃BNO



Scheme 1. Schematic illustration of the synthesis of the novel green dopant, 3-CF₃BNO.

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