

# Synthesis and characterization of homoleptic triply cyclometalated iridium (III) complex containing 6-(pyridin-2-yl)isoquinoline moiety for solution-processable orange-phosphorescent organic light-emitting diodes

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## ABSTRACT

The design and synthesis of a novel homoleptic triply cyclometalated iridium(III) complex containing the 6-(pyridin-2-yl)isoquinoline moiety [Ir(pyiq)<sub>3</sub>] was demonstrated for the first time. The performance of a phosphorescent organic light emitting diode (PHOLED) based on Ir(pyiq)<sub>3</sub> is described with adoption of tris(4-carbazoyl-9-ylphenyl)amine (TCTA) and 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) which are co-host materials showing excellent compatibilities as well as CBP host with the prepared phosphorescent dopants. The photoluminescence (PL) of Ir(pyiq)<sub>3</sub> produced orange emission with maximum emission peak at 583 nm. The highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital energy (LUMO) levels of Ir(pyiq)<sub>3</sub> were −5.41 eV and −3.20 eV. An optimized solution-processed device doped with Ir(pyiq)<sub>3</sub> had a maximum external quantum efficiency (EQE) of 8.71% and a maximum current efficiency (CE) of 22.51 cd/A. This correspond to 17% higher efficiency than that with bis(2-phenylbenzothiazolato-N,C<sup>2'</sup>) iridium(acetylacetonate) (bt)<sub>2</sub>Ir(acac), which is commonly used in orange PHOLEDs. This study rationalizes the promising application of new trimeric organometallic complex that possesses isoquinoline and pyridine in solution-processed PHOLEDs with good quantum and power efficiency.

## 1. Introduction

The organic light-emitting diode (OLED) is a key light source for display technologies and solid-state illumination devices due to its compelling properties, namely low cost, light weight, wide viewing angle, fast response time, good color saturation and resolution, and satisfactory power efficiency [1]. It is considered as an energy-saving and ecofriendly next-generation technology. Tang and VanSlyke first constructed a practical OLED device in 1987 using tris(8-hydroxyquinoline)aluminum(III) (Alq<sub>3</sub>) [2]. A tremendous amount of effort followed to improve electroluminescent emission efficiency, response performance, ease of device fabrication, and low driving

voltage load [3]. *Inter alia*, the development of organic materials as the emitting elements has been progressively studied in terms of their synthesis, and many industrial and academic research groups have designed and characterized novel emitters displaying high external quantum efficiencies (EQEs) and power efficiencies [4].

Phosphorescent organic light-emitting diodes (PHOLEDs) are based on the principle of phosphorescence, which enables an OLED to convert electrical energy into light in a highly efficient manner. Theoretically, the internal quantum efficiency can approach 100% by capturing both singlet and triplet excitons [5]. Forrest et al. introduced 2,3,7,8,12,13,17,18-octaethyl-21H,23H-porphine platinum(II) (PtOEP) as the first phosphor-doped OLED in 1998 [6]. Since then, a wide variety of

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phosphorescent emitters based on organometallic structures have been developed [7]. Organometallic complexes, which consist of centrally located metals and peripheral organic compounds, called ligands, exhibit spin-orbit coupling that facilitates intersystem crossing between singlet and triplet states with the aid of a central heavy metal atom. Such complexes generally have relatively high phosphorescence efficiency due to the strong radiative  $n-\pi^*$ ,  $\pi-\pi^*$  transition moment and metal-to-ligand charge transfer (MLCT) [8]. Iridium complexed with various chelating ligands is often strongly luminescent [9]. The Ir(III) complexes most extensively studied as efficient phosphor dopants are *fac*-Ir(ppy)<sub>3</sub> and related complexes [10]. Numerous investigations have reported that certain cyclometalating ligands, the installation of electron-withdrawing or electron-releasing groups on phenyl or pyridine rings, and the inclusion of appropriate ancillary ligands can significantly influence passive emissive properties and photoluminescent efficiency [11].

Two methods have been mainly used to prepare amorphous films for OLED devices. The classical approach is vapor deposition via a thermal evaporation process under high vacuum. The ideal multilayer structure has high thermal stability, high purity, and significant horizontal molecular orientation [12]. However, limitations to this approach include the high fabrication cost and, especially, the difficulty in fabricating large-area devices. The second method involves solution processing, which enables low-cost manufacture of high-performance large-area applications [13]. To construct PHOLEDs by the solution process, a high doping concentration of the phosphorescent dopant is needed to mitigate triplet exciton release onto the host; this has driven research to improve dopant solubility in organic solvents [14]. Although Ir(ppy)<sub>3</sub> has somewhat low solubility in common solvents such as toluene, tetrahydrofuran (THF), dichloromethane (DCM), and chloroform, some Ir(III) complexes, notably bis(4,6-(difluorophenyl)pyridinato-N,C20picolate)iridium(III) (Flrpic), tris(2-(*p*-tolyl)pyridine)iridium(III) (Ir(mppy)<sub>3</sub>), tris(1-phenylisoquinoline-C2,N)iridium(III) (Ir(piq)<sub>3</sub>), and bis(1-phenylisoquinoline)(acetylacetonate)iridium(III) (Ir(piq)<sub>2</sub>(acac)), have higher solubilities [15,16].

In this work, a novel homoleptic triply cyclometalated iridium(III) complex, Ir(piq)<sub>3</sub> was designed and prepared as the orange-red-phosphorescent emitter for the first time. This molecule features three 6-(pyridin-2-yl)isoquinolines, covalently adducts between the carbon labeled number 6 of isoquinoline ring and the carbon labeled number 2 of pyridine ring. The center atom iridium is coordinated to every 7-positioned carbons of isoquinolines and nitrogens of pyridines affording the significant structural stability and enough solubility under solution-processed device construction. With the molecule in hand, upon the fabrication of solution-processed PHOLED, optimizations by finely tuning the ratio of the co-host, 4,4',4''-tris(*N*-carbazolyl)-triphenylamine (TCTA): 2,2,2-(1,3,5-benzenetriyl)tris-[1-phenyl-1H-benzimidazole] (TPBI), annealing time and temperature were performed. Furthermore, the thermal stability, electrochemical features, and photophysical properties of Ir(piq)<sub>3</sub> doped OLED were investigated, and compared to those of the well-known Ir(III) complex, (bt)<sub>2</sub>Ir(acac) [17].

## 2. Results and discussion

### 2.1. Synthesis and characterization

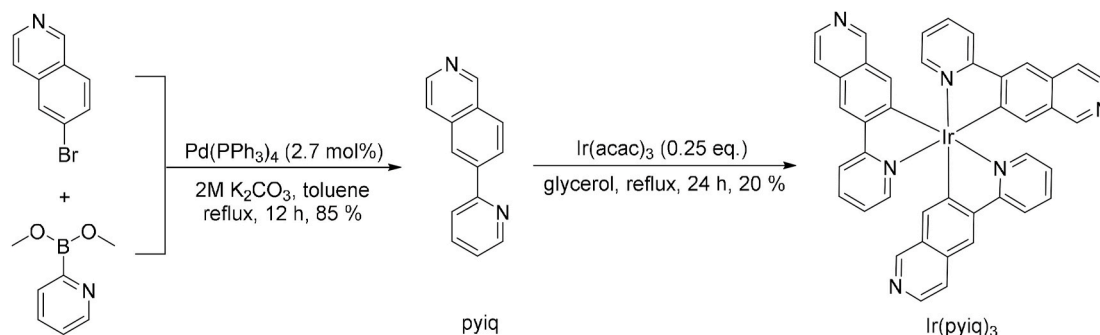
The designed Ir(III) complex, Ir(piq)<sub>3</sub> were synthesized through the route depicted in Scheme 1. The cyclometalating ligand, piq was prepared by Suzuki coupling reaction [17] which make connect two aryl rings, 6-bromoisoquinoline and dimethyl pyridine-2-yl boronate, in the presence of Pd(0) catalyst with good chemical yield of 85% as a white solid. This adduct molecule was run under a ligand exchanging reaction using Ir(acac)<sub>3</sub>, and trimerization of ligands was performed in an expected manner using glycerol solvent [18]. Protons and carbon atoms of the synthesized molecules were assigned using their respective <sup>1</sup>H and <sup>13</sup>C nuclear magnetic resonance and mass spectra.

### 2.2. Device fabrication

The solution-processed orange PHOLED was fabricated as follows. Indium-tin oxide (ITO)-patterned glass substrates were sequentially cleaned with acetone and isopropanol for 15 min each using an ultrasonic bath. After drying the ITO-patterned substrates, the surface was treated with ultra-violet (UV) ozone for 15 min. Then, a 60-nm-thick polymeric hole injection layer having gradient work-function (GraHIL; a blend of poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT:PSS) and a perfluorinated ionomer (PFI) with a weight ratio of 1:3.6) was spin-coated and baked at 150 °C for 30 min. A 50-nm-thick emitting layers (EMLs), which were composed of 10% Ir(piq)<sub>3</sub>, bis(2-phenylbenzothiazolato-N,C [2']iridium(acetylacetonate) ((bt)<sub>2</sub>Ir(acac)) as orange phosphorescent dopants and 4,4'-bis(9-carbazolyl)-biphenyl (CBP) as the single host or TCTA:TPBI (at weight ratio and concentration of 0.6:0.4 and 0.25 wt%, respectively) as the mixed host, were spin-coated from THF (SAMCHUN Chemical Co.). After the deposition of EMLs, 50-nm-thick TPBI as an electron transporting layer, LiF as an electron injection layer, and Al as a cathode were sequentially thermally-deposited onto the EMLs under high vacuum condition ( $\sim 10^{-7}$  Torr). Finally, the fabricated devices were encapsulated using a hollow glass and UV-epoxy resin. The device structure of the solution-processed orange PHOLEDs and the chemical structures of the host materials are shown in Fig. 1.

### 2.3. Photophysical, electrochemical and thermal properties

As shown in Fig. 2, the maximum ultraviolet (UV)-visible (Vis) absorption of Ir(piq)<sub>3</sub> was shown at 254 nm in an anhydrous dichloromethane solution. The weak shoulder absorption band at about 320 nm was assigned to ligand-centered (LC) transitions with spin-allowed  $\pi-\pi^*$  character. Quite weak but overtly exist light absorption at around 450 nm was attributed to spin-allowed and spin-forbidden metal-to-ligand charge transfer (MLCT) transitions within the organometallic complex. Photoluminescence (PL) has demonstrated the maximum emission at the wavelength of 583 nm, at which orange color is released. It is known



Scheme 1. Synthetic route for the organometallic complex, Ir(piq)<sub>3</sub>.

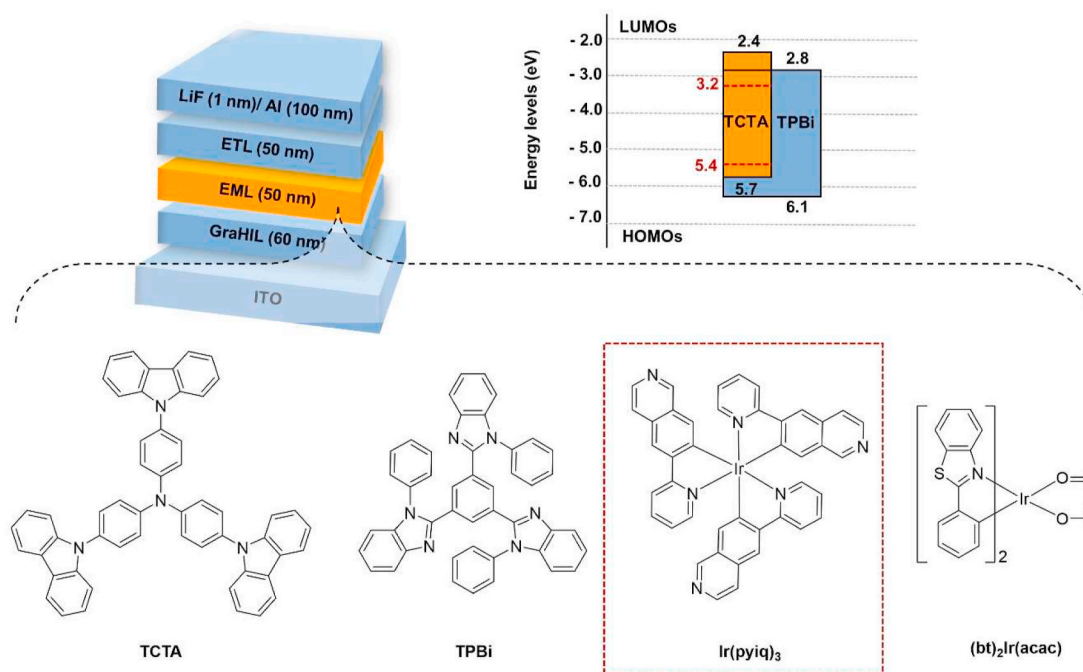


Fig. 1. Device structure of solution-processed PHOLEDs, energy levels of materials, and chemical structures of host materials and dopant molecules.

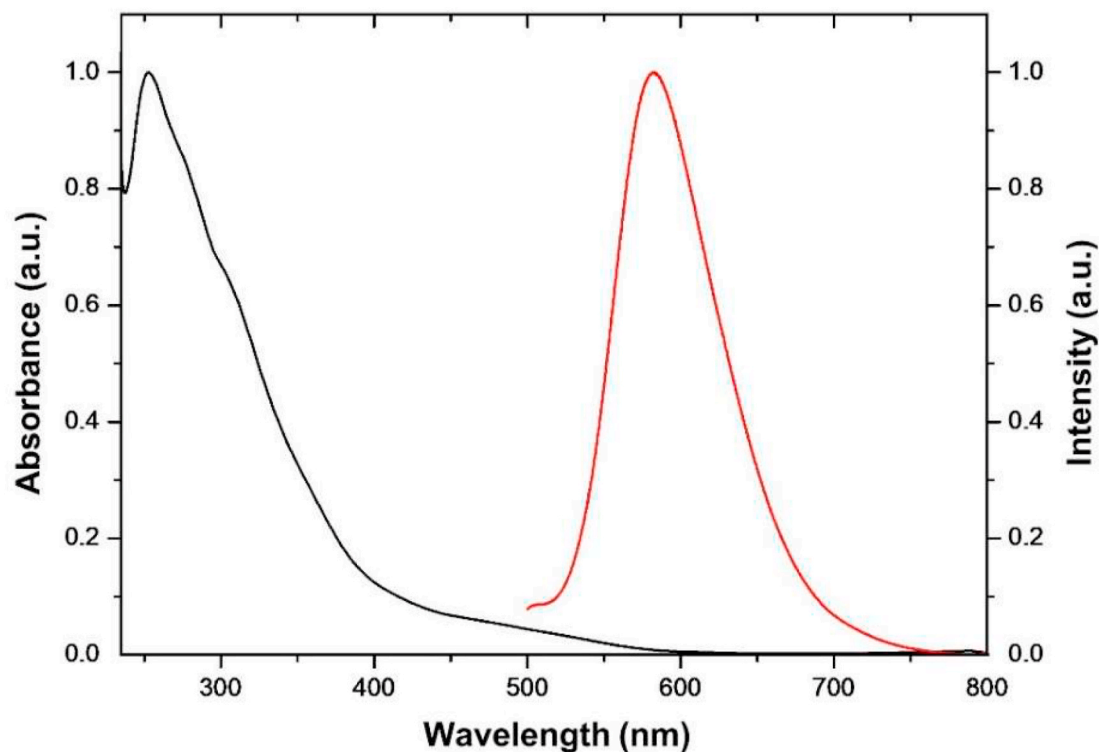


Fig. 2. UV-visible absorption and PL spectra of Ir(pyiq)<sub>3</sub> in CH<sub>2</sub>Cl<sub>2</sub> solution.

that although the standard tree colors, R (red), G (green), and B (blue) is needed for full color display with a high color purity, yellow and/or orange emitters are quite beneficial for white light to cover a wide spectral region as well as lighting application [19,20]. The band gap was obtained to be 2.21 eV was calculated from the onset of UV-vis absorption (561 nm). The full width at half maximum (FWHM) of 82 nm accounts for favorable structure rigidity of two kinds of aryl rings in Ir(pyiq)<sub>3</sub>. Table 1 lists the values of other photophysical, thermal, and

electrochemical data.

The thermal behavior of Ir(pyiq)<sub>3</sub> was investigated by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) under nitrogen atmosphere as shown in Fig. 3. The thermal-stability data that reveals the value of 5% weight-reduction temperatures ( $\Delta T_{5\%}$ ) as 442 °C means the new complex has an excellent thermal stability. According to the heat flow chart no crystallization temperature ( $T_c$ ) and melting transition temperature ( $T_m$ ) was shown at all while the

**Table 1**  
Photophysical, thermal and electrochemical information of Ir(pyiq)<sub>3</sub>.

UV, $\lambda_{\text{abs}}$ (nm)	PL, $\lambda_{\text{em}}$ (nm)	FWHM (nm)	$\Delta T_{5\%}$ (°C)	HOMO (eV)	LUMO (eV)	Eg (eV)
254	583	82	442	−5.41	−3.20	2.21

PL: photoluminescence, FWHM: The full width half maxima,  $\Delta T_{5\%}$ : 5% weight-reduction temperature, HOMO: the highest occupied molecular orbital, LUMO: the lowest unoccupied molecular orbital, Eg: the energy band gap.

glass transition temperature ( $T_g$ ) may seemed to be observed at around 180 °C, the Ir(III) complex was enough to be regarded to maintain its amorphous solid state worthily. The obvious thermal data above means the newly-prepared Ir(III) complex is quite temperature endurable favorable for used as a dopant in OLED. The highest-occupied molecular orbital (HOMO) energy level was obtained from oxidation onset of cyclic voltammetry (CV) and the lowest-unoccupied molecular orbital (LUMO) energy value was calculated by HOMO and band gap (Fig. S6).

To investigate the photophysical properties of Ir(pyiq)<sub>3</sub>, the photoluminescence quantum yield (PLQY) and PL lifetime were measured by synthesizing 0.25 wt% Ir(pyiq)<sub>3</sub>:dichloromethane (DCM) solution. The measurements were performed at room temperature by using FluoroTime300 spectrometer with a laser excitation wavelength of 405 nm. The measured PL lifetime and PLQY were 0.32  $\mu$ s and 29.4%, respectively (Table 2, Fig. 4). The relatively shorter PL lifetime in the micro-second regime than the milliseconds regime shows that Ir(pyiq)<sub>3</sub>, a homoleptic triply cyclometalated Ir complex, has strong spin-orbital coupling which enables to use triplet excitons (Fig. 4). Moreover, the measured PLQY of Ir(pyiq)<sub>3</sub> (PLQY<sub>Ir(pyiq)3</sub> = 29.4%) was improved more than 13% than that of Bt<sub>2</sub>Ir(acac) (PLQY<sub>Bt2Ir(acac)</sub> = 26%) that is a commercially-available orange-phosphorescent organic dopant [15]. Therefore, we confirmed that Ir(pyiq)<sub>3</sub> has a remarkable photophysical properties as an orange-phosphorescent emitter.

#### 2.4. Electroluminescent properties

We fabricated simple-structured and orange PHOLEDs to evaluate the electroluminescence (EL) properties of Ir(pyiq)<sub>3</sub> as a phosphorescent orange dopant based on a solution process using CBP as a single host as well as TCTA:TPBI as a mixed-host for EMLs. Moreover, the simple device structure is advantageous to achieve high-efficiency solution-processed OLEDs because the continuous solution processing can damage an underlying layer and thus depositing organic multilayer is one of important issues in the fabrication of solution-processed OLEDs. The polymeric GraHIL tolerates subsequent solution processing using an organic solvent, and its high work function (~5.95 eV) facilitates the hole injection from the ITO anode to EMLs. Both CBP and TCTA:TPBI can behavior as bipolar charge transporting hosts, but energy levels of

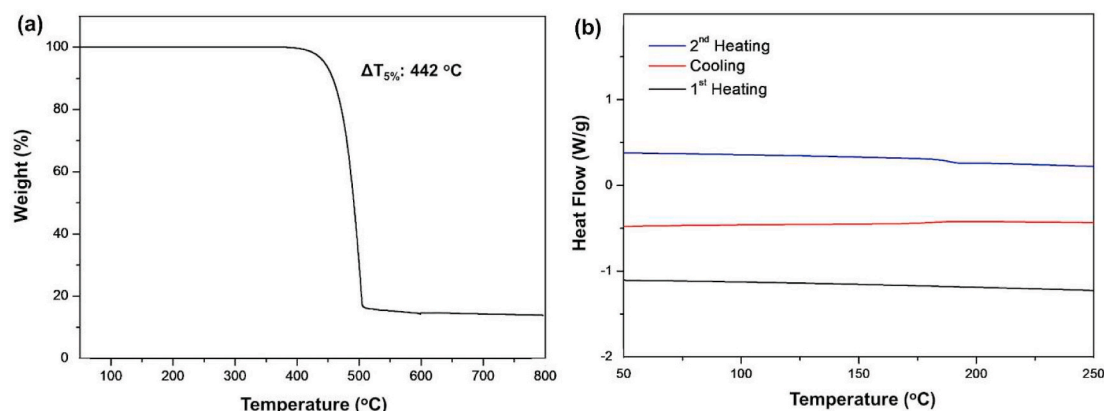
each host are different. Charge injection into the mixed host EML can be more efficient than the single host EML, because the hole injection energy barrier from GraHIL and electron injection barrier from the TPBI ETL are lower in the mixed host than in the single CBP host (Fig. 5a). Consequently, TCTA:TPBI:Ir(pyiq)<sub>3</sub> displayed higher maximum current efficiency (CE, 22.51 cd/A) than CBP:Ir(pyiq)<sub>3</sub> (19.95 cd/A) (Fig. 5b, Table S1). On the other hand, the maximum EQE value of Ir(pyiq)<sub>3</sub>-based PHOLEDs was achieved when the CBP single host was used (8.71% for CBP, 8.46% for TCTA:TPBI) at low current density under 0.01 mA/cm<sup>2</sup> (Fig. 5c). This behavior likely originated from the blue-shifted EL spectrum of CBP:Ir(pyiq)<sub>3</sub> compared to that of TCTA:TPBI:Ir(pyiq)<sub>3</sub> due to different recombination position in each EMLs (Fig. 5d). However, in the operational level of the current density above 0.1 mA/cm<sup>2</sup>, the EQE of TCTA:TPBI:Ir(pyiq)<sub>3</sub> maintained higher value than that of CBP:Ir(pyiq)<sub>3</sub>. Finally, we compared the EL properties of Ir(pyiq)<sub>3</sub> with a conventional orange dopant, (bt)<sub>2</sub>Ir(acac) in the TCTA:TPBI mixed host. The CE and EQE of TCTA:TPBI:(bt)<sub>2</sub>Ir(acac) showed 19.28 cd/A and 7.11%, which are lower than that of TCTA:TPBI:Ir(pyiq)<sub>3</sub>. Therefore, we confirmed that a novel homoleptic triply cyclometalated iridium(III) complex, Ir(pyiq)<sub>3</sub>, can be effectively used for the solution-processed PHOLEDs and exceed the EQE of conventional phosphorescent orange dopant [16].

#### 3. Conclusion

We designed and synthesized a homoleptic triply cyclometalated iridium(III) complex containing 6-(pyridin-2-yl)isoquinoline moiety, specifically, [Ir(pyiq)<sub>3</sub>]. The thermal stability, electrochemical properties, and photophysical behaviors of Ir(pyiq)<sub>3</sub> doped OLED were studied. Ir(pyiq)<sub>3</sub> produced orange emission with a maximum emission peak at 583 nm, which suggested that this novel emitter is a good alternative to the conventional emitting materials for white OLED displays. The highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital energy (LUMO) levels of Ir(pyiq)<sub>3</sub> were calculated as −5.41 eV and −3.20 eV respectively. By optimization the fabrication of the solution-processed device in which Ir(pyiq)<sub>3</sub> was doped in the EML, a maximum EQE of 8.71% and a maximum CE of 22.51 cd/A were realized. This achievement shows the amelioration in efficiency of 17% compared to the performance of (bt)<sub>2</sub>Ir(acac) that is one of the conventional orange phosphorescent OLED.

**Table 2**  
Photophysical properties of Ir(pyiq)<sub>3</sub>. PL lifetime and PL quantum yield were measured at room temperature by using FluoroTime300 spectrometer.

	PL lifetime ( $\mu$ s)	PLQY (%)
Ir(pyiq) <sub>3</sub>	0.32	29.4%



**Fig. 3.** The (a) thermo-gravimetric analysis (TGA) and (b) differential scanning calorimetry (DSC) curves of Ir(pyiq)<sub>3</sub>.



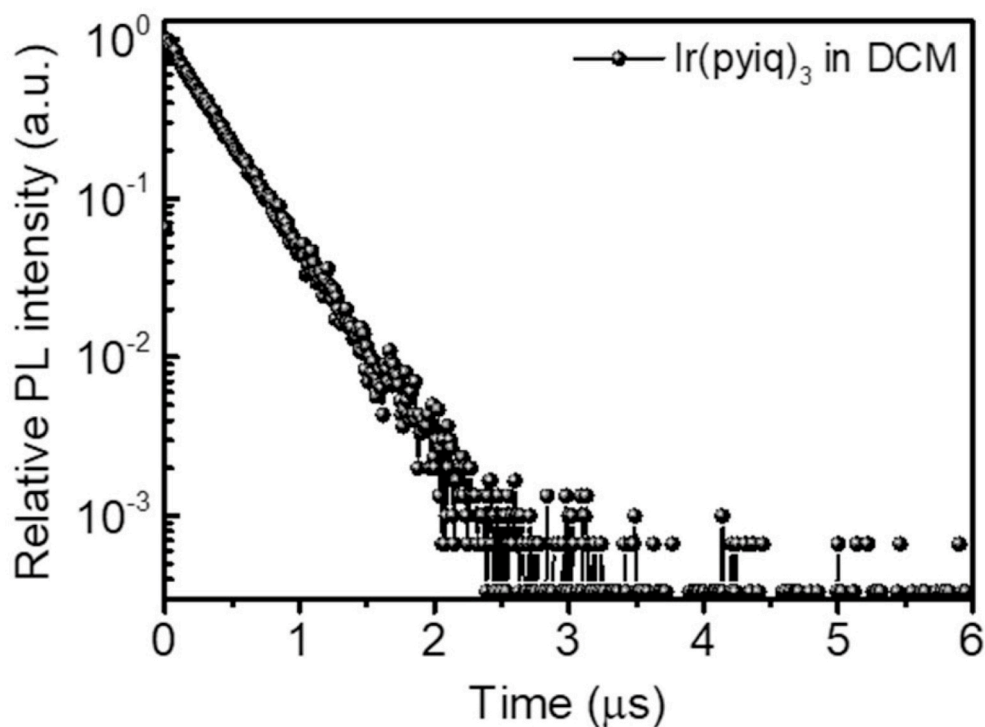


Fig. 4. Photoluminescence lifetime decay profile of Ir(pyiq)<sub>3</sub> at excitation wavelength of 405 nm.

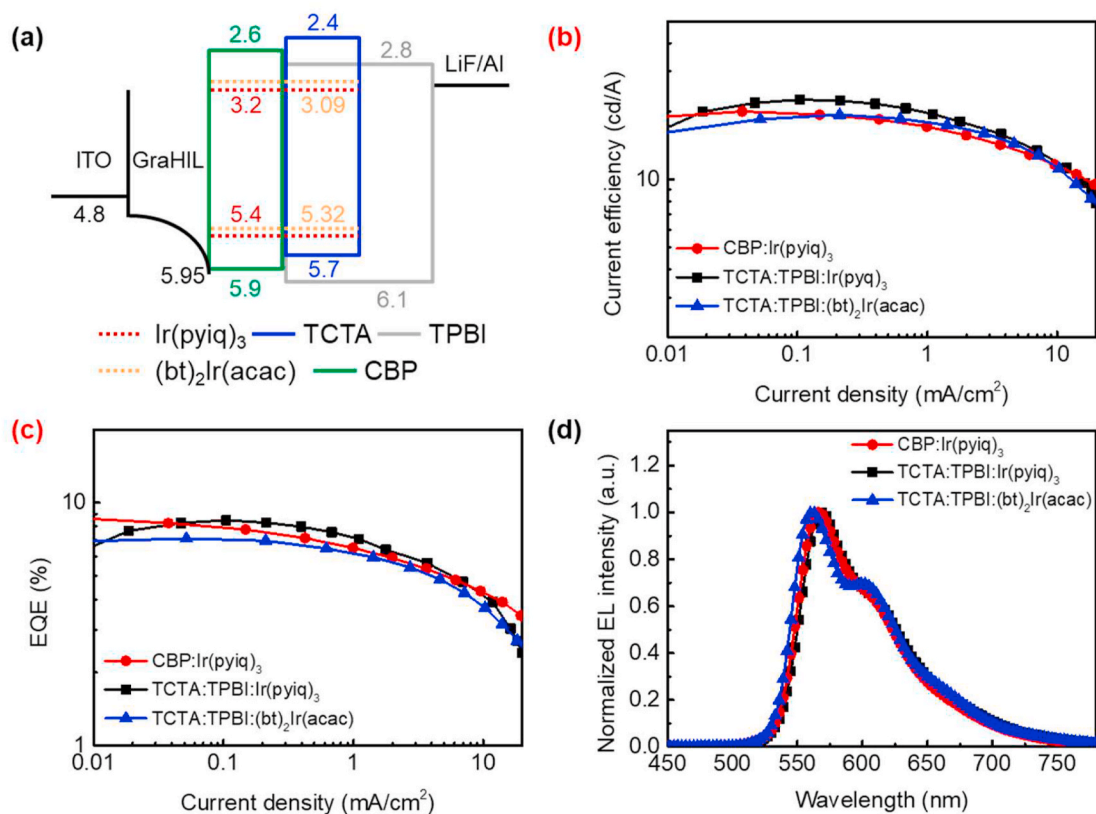


Fig. 5. (a) Energy level diagram of fabricated orange PHOLEDs. Device characteristics of (b) current efficiency (CE) versus current density, (c) external quantum efficiency (EQE) versus current density, and (d) electroluminescence (EL) spectra of each PHOLEDs.

## Author statement

**Min Woo Ha:** Investigation and Writing Original Draft. **Min-Ho Park:** Investigation and Writing Original Draft. **Ja Young Hwang:** Formal analysis. **Jingwan Kim:** Formal analysis. **Dong-Heok Kim:** Formal analysis. **Tae-Woo Lee:** Writing Review and Editing, Supervision. **Yun-Hi Kim:** Conceptualization, Writing Review and Editing, Supervision

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.dyepig.2020.108880>.

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