Solution-processed sky-blue phosphorescent organic light-emitting diodes based on 2-(5,9-dioxa-13b-boranaphtho[3,2,1-de]anthracene-8-yl)-4-(trimethylsilyl)pyridine chelated iridium complex

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ABSTRACT
A new 2-(5,9-dioxa-13b-boranaphtho[3,2,1-de]anthracene-8-yl)-4-(trimethylsilyl)pyridine-based iridium complex was synthesized for efficient solution-processed sky-blue phosphorescent organic light-emitting diodes (PhOLEDs). The effect of dioxaboranaphthoanthracene instead of phenyl with the electron-accepting group, as well as the bulky pyridine with the trimethylsilyl group, on the ligand was investigated. The new dopant was found to have an extremely high photoluminescence quantum yield of 94% when doped in an emissive layer. As a result, the solution-processed blue PhOLED consisting of a simple structure without any interlayer exhibited remarkable light-emitting performance with an external quantum efficiency of 8.93% and a current efficiency of 23.56 cd/A.

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Introduction
Light-emitting transition metal complexes have been widely used as emitting dopants in the fields of organic light-emitting diodes (OLEDs) [1,2]. Cyclometalated iridium(III) complexes are one of the most thoroughly studied classes of compounds for these applications due to their excellent thermal stability and photostability, relatively short phosphorescence lifetime, easy color tuning, and high photoluminescence quantum yield [3–8]. This complex has two main structural classes: a homocomplex of type [Ir(CN)3] (CN = cyclometalated ligand), particularly in a fac geometry, and the general formula [Ir(CN)2(LL')]n (LL' = ancillary ligand, n = −1, 0, +1). Color tuning is most often achieved by altering the structure of the cyclometalated ligand; however, ancillary ligands in the heteroleptic complex can sometimes affect the emission color [9–11]. The combination of cyclometalating and ancillary ligands also characterizes the emission T1 state, which is mostly a combination of ligand center (3LC or 3π*) and intermetallic charge transfer (3MLCT or 3dπ*) mixing. Although the structure–property relationship of cyclometalated iridium complexes has been extensively studied, an ongoing challenge in this field is designing molecules that ensure stable and efficient deep-blue emission [11–13].

The most common design rule for blue-light emission of iridium complexes is to introduce an electron donor group at the pyridine group and an electron-withdrawing group at the phenyl group. Since the electron-withdrawing group in the phenyl moiety lowers the highest occupied molecular orbital (HOMO) level, a phenyl part with various electron-withdrawing groups introduced, such as 2,4-difluorophenyl, 3-trifluorocarboxylated-2,4-difluorophenyl, 3-trifluorocarboxylated-2,4-difluorophenyl, 3-trifluoromethyl-2,4-difluorophenyl, 3-nitro-2,4-difluorophenyl, 3-pyridyl-2,4-difluorophenyl, was designed [14–24]. A trimethylsilyl group was recently introduced in the pyridine part, which increased the bandgap of the emitter and increased the horizontal...
dipole orientation (θ) of the Ir complexes, thereby leading to a highly efficient blue emission [24]. In addition, further research has been conducted on solution-based processes instead of the vacuum-deposited process in order to realize cost-effective and large-area mass production of organic electronics [25–29]. These previously reported literatures are exemplary in that they not only demonstrate the great potential of soluble OLEDs, but also the undeniable role of highly efficient electrophosphorescent emitters in achieving high-performance OLEDs in the realization of low-cost, large-area, and inkjet-printed electronics.

In this study, a new 2-(5,9-dioxa-13b-boranaphtho[3,2,1-de]anthracene-8-yl)−4-(trimethylsilyl)pyridine-based iridium complex, (BOPySi)2IrPic, was designed and synthesized for high-efficiency solution-processed sky-blue phosphorescent OLEDs (PhOLEDs). To lower the HOMO energy level, the 2-(5,9-dioxa-13b-boranaphtho[3,2,1-de]anthracene-8-yl) group was introduced instead of the phenyl part with general electron acceptor groups. Furthermore, the introduction of the trimethylsilyl group has led to the increase in the bandgap and horizontal dipole orientation. It is also advantageous for easily dissolving the synthesized dopant in common organic solvents because of the trimethylsilyl functional group in the boron containing ligand. The solution-processed sky-blue PhOLEDs with a (BOPySi)2IrPic emitter-doped emissive layer (EML) exhibited a peak external quantum efficiency (EQE) of 8.93% with a current efficiency (CE) of 23.56 cd/A.

### Experimental

Figure 1 (a) demonstrated solution-processed PhOLEDs based on the two new emitters that were fabricated on glass substrates with a structure of indium tin oxide (ITO, 150 nm) as the bottom anode, PEDOT:PSS (40 nm) as HIL, mCP:TSP01: (BOPySi)2IrPic (1:1:20 wt%, 40 nm) as single EML, BmPyPB (x nm) as ETL, LiF (1 nm) as the electron injection layer (EIL), and aluminum (Al) (100 nm) as the top cathode. A mixed-host layer consisting of a hole-transporting host, 1,3-bis(N-carbazolyl) benzene (mCP), and an electron-transporting host, diphenyl[4-(triphenylsilyl)phenyl]phosphine oxide (TSP01), was doped with (BOPySi)2IrPic as an emitting layer (EML). Then, 40-nm-thick hole injection layers (HILs) of poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) and electron transfer layers (ETLs) of 1,3-bis(3,5-dipyrid-3-yl-phenyl)benzene (BmPyPB) with thicknesses of 30, 40, and 50 nm were fabricated. The mixed-host mCP:TSP01 (1:1) and dopant of (BOPySi)2IrPic were dissolved in the toluene solvent. The total concentration of the EML solution was 1.0 wt%. The detailed device fabrication and encapsulation methods were listed in the supporting information.

### Results and discussion

The synthetic routes and chemical structures of (BOPySi)2IrPic are shown in Scheme 1. BOPySi ligand was synthesized by using the Suzuki coupling reaction. Cyclometalated iridium m-chloro-bridged dimer was prepared by using the well-known Nonoyama reaction between the ligand and IrCl3 ·xH2O in a mixture of water and 2-ethoxyethanol (5 mL: 15 mL). The new (BOPySi)2IrPic sky-blue Ir (III) complex was obtained via the reaction of the μ-chloro-bridged dimer with 2-picolinic acid in 2-ethoxyethanol. The structure of the intermediates and new iridium complexes was confirmed by 1H-NMR, 13C-NMR, and HRMS measurements. The supporting information contains the details of the synthesis and characterizations.

The photophysical properties of the newly synthesized sky-blue emitters, (BOPySi)2IrPic, in dilute chloroform solutions were examined via the UV-vis absorption and photoluminescence (PL) spectra, as shown in Figure 2. Meanwhile, the photophysical data of (BOPySi)2IrPic are summarized in Table 1. The absorption maximums of (BOPySi)2IrPic are approximately 255, 349, and 410 nm. It is known that 255 nm can be matched to the

![Figure 1](image-url)
\( \pi \rightarrow \pi^* \) transition of the cyclometalated ligand, while 349 nm belongs to the singlet metal ligand charge transfer transition \(^{1}\text{MLCT}\) transition. Moreover, the weak 410 nm may belong to the triplet metal ligand charge transfer transition \(^{3}\text{MLCT}\). Photoluminescence was observed at approximately 479 nm with a 509-nm shoulder, which is 4-nm red-shifted compared with bis(4,6-difluorophenylpyridinato-N,C2)picolinatoiridium (FIrpic). \( T_1 \) was calculated to be almost 2.77 eV from the onset of the low-temperature PL measurements.

In addition, the thermal properties of the synthesized dopant were characterized by thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC) measurements were conducted under an \( \text{N}_2 \)-filled atmosphere (see Fig. S1 and Table 1). The TGA results revealed that \((\text{BOPySi})_2\text{IrPic}\) has remarkable thermal stability, with a 5% weight loss temperature \((\Delta T_{5\%})\) of more than 430°C. However, the DSC results did not reveal the temperatures of the glass transition \((T_g)\), melting \((T_m)\), and crystallization \((T_c)\) of the materials.

The HOMO and the lowest unoccupied molecular orbital (LUMO) energy level of \((\text{BOPySi})_2\text{IrPic}\) could be estimated from cyclic voltammograms and the optical bandgap from the absorption edge, as shown in Fig. S2. In addition, the HOMO energy level of \((\text{BOPySi})_2\text{IrPic}\) was calculated to be \(-5.5\) eV according to the equation of \(E_{\text{HOMO}}\) (eV) = \(-4.84 - [E_{\text{ox, onset}} - E_{1/2}(\text{ferrocene})]\). The value of the energy gap \(E_g\) was obtained by using the UV-vis absorption spectrum edge \(\lambda_{\text{a, edge}}\) based on the formula of \(E_g = 1240/\lambda_{\text{a, edge}}\). The calculated optical bandgap of \((\text{BOPySi})_2\text{IrPic}\) is 2.90 eV. The LUMO energy level was estimated from the difference between the optical bandgap of 2.9 eV and the HOMO energy level of \(-5.5\) eV by using the equation \(E_{\text{LUMO}} = E_g + E_{\text{HOMO}}\).

The LUMO energy level of \((\text{BOPySi})_2\text{IrPic}\) calculated by using this equation was \(-2.60\) eV, as listed in Table 1.

In order to achieve high light-emitting performance, the choice of the host material is important, particularly the high photoluminescent quantum yield (PLQY) of the emitter-doped host layer. To meet this requirement, the dependency of the PLQY on the host variations was tested. In particular, mCP, TSPO1, and a mixture of mCP and TSPO1 were tested as host materials, which have been known to be good host materials for solution-processed blue PhOLEDs due to their high triplet energy levels and good charge transfer properties. It was found that the \((\text{BOPySi})_2\text{IrPic}\) emitter had much higher PLQY values when doped in a mixed-host layer consisting of a hole-transporting host (mCP) and an electron-transporting host (TSPO1) than with a single host structure. The absolute PLQYs of \((\text{BOPySi})_2\text{IrPic}\) were measured as 90%, 31%, and 94%, respectively, for 20% doping mCP, mCBP, and mCP: TSPO1(1:1) films. As

| Compound      | UV \(\lambda_{\text{max}}\) (nm) | PL \(\lambda_{\text{max}}\) (nm) | FWHM (nm) | \(T_d,5\%\) (°C) | \(T_g\) (°C) | \(T_m\) (°C) | HOMO (eV) | LUMO (eV) | \(E_g\) (eV) | \(T_1\)   |
|---------------|---------------------------------|---------------------------------|-----------|-----------------|------------|------------|-----------|-----------|-------------|---------|
| \((\text{BOPySi})_2\text{IrPic}\) | 255, 349                       | 479, 509                        | 46.4      | 430             | –          | –          | 5.5       | 2.6       | 2.9         | 2.77    |

Figure 2. UV-vis absorption and PL spectrum of \((\text{BOPySi})_2\text{IrPic}\).

Figure 3. Current density \((J)\)–voltage \((V)\)–luminance \((L)\) characteristics of the solution-processed PhOLED.

Figure 4. Normalized electroluminescence (EL) spectra.
Table 2. Device performances of solution processed OLEDs in this study

| dopant     | ETL Thickness | EQE (%) | CE (cd/A) | CIE coordinates (x, y) |
|------------|---------------|---------|-----------|------------------------|
| (BOPySi)_2IrPic | 30nm          | 5.58    | 14.50     | (0.20, 0.53)           |
|            | 40nm          | 8.09    | 21.16     | (0.21, 0.53)           |
|            | 50nm          | 8.93    | 23.56     | (0.22, 0.53)           |

A result, solution-processed OLEDs were fabricated with this mCP:TSPO1 mixed-host structure.

The current density (J)–voltage (V)–luminance (L) characteristics and normalized electroluminescence (EL) spectra of two (BOPySi)_2IrPic-doped sky-blue PhOLEDs with 50-nm-thick BmPyPB ETLs are shown in Figures 3 and 4, respectively. The device performances of the OLEDs used in this study, including key light-emitting performance parameters and EL emission characteristics, are summarized in Table 2. Solution-processed PhOLEDs showed low turn-on voltages at approximately 4.0 V at 1 cd/m², which could be explained by relatively small injection barriers from both the ETL to the EML and the HTL to the EML, as shown in the band diagram of Figure 1. The low injection barrier can be attributed to the mCP:TSPO1 mixed host, as well as the adequate HOMO/LUMO energy level of the dopants.

In Figure 4, the EL spectra of the PhOLEDs showed peak emission at approximately 489 nm, which is slightly red-shifted compared with the (BOPySi)_2IrPic PL emission due to the strong intermolecular interactions and the optimization of optical length. Shoulder EL emission at approximately 516 nm was found and verified in the PL study. The color coordinates of (BOPySi)_2IrPic with 50-nm-thick BmPyPB ETL were (0.22, 0.53).

The EQE and CE values as a function of the luminance are given in Figure 5 and Table 2. Based on the host combination, the PLQY value of (BOPySi)_2IrPic in an mCP and TSPO1 mixed host was quite high at 94%, as shown in Table 3. This result could improve the light-emitting performances of solution-processed devices. In general, the solution-processed PhOLEDs with (BOPySi)_2IrPic emitter showed high efficiencies in terms of EQE and CE, even though the device structure is very simple, without any interlayer in the HIL–EML and EML–ETL interfaces. The solution-processed (BOPySi)_2IrPic-based PhOLEDs with 30-nm, 40-nm, and 50-nm BmPyPB ETL thicknesses showed 5.58%, 8.09%, and 8.93% EQE values, respectively. In addition, the (BOPySi)_2IrPic device with a 50-nm-thick BmPyPB ETL device showed the highest CE (23.56 cd/A). It is possible to achieve high-performance solution-processed OLEDs with a very simple structure by optimizing ETL thickness, selecting an appropriate host, and developing a new dopant combination. As illustrated in Fig. S3, we also found that the light-emitting performances of (BOPySi)_2IrPic emitter-based device in this study show highly improved efficiencies compared to those of FIrpic-based OLED, which is the most well-known reference phosphorescent sky-blue dopant.

Table 3. PLQY values of (BOPySi)_2IrPic doped host variations

| PLQY (%) | mCP host | mCBP host | mCP:TSPO1 host |
|----------|----------|-----------|----------------|
| (BOPySi)_2IrPic | 90       | 31        | 94             |

Figure 5. (a) External quantum efficiency (EQE) and (b) current efficiency (CE) — luminance characteristics of the device designed.

Conclusion

A new 2-(5,9-dioxo-13b-boranaphtho[3,2,1-de]anthracene-8-yl) electron-acceptor-group-based iridium complex was designed and synthesized for solution-processed sky-blue PhOLEDs. The new dopant doped in a mixed host of mCP:TSPO1 showed an extremely high PLQY of 94%. The resultant solution-processed sky-blue PhOLED with a very simple device structure exhibited a high CE of 23.56 cd/A and EQE of 8.93% through the optimization of the doping concentration and ETL thickness. Therefore, a new molecular design was proposed in order to
develop high-efficiency printing-processed phosphorescent OLEDs.

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**Disclosure statement**

No potential conflict of interest was reported by the author(s).

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