



A thermally stable imidazole type ligand based Be complex as a triplet host material of green phosphorescent organic light emitting diodes



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ABSTRACT

A 2-(4,5-dimethyl-1-phenyl-imidazol-2-yl)phenol ligand based Be complex, beryllium 2-(4,5-dimethyl-1-phenyl-imidazol-2-yl)phenolate (BePhIm), was investigated as a thermally stable host material of green phosphorescent organic light-emitting diodes (PHOLEDs). BePhIm was developed as a host material to have high triplet energy, thermal stability and balanced charge density. The triple energy of BePhIm was 2.73 eV and the glass transition temperature was 181 °C. Maximum quantum efficiency of the BePhIm based green PHOLEDs was 25.9%, which is better than any other quantum efficiency reported in green PHOLED using Be complexes as the host material.

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1. Introduction

The quantum efficiency (QE) of phosphorescent organic light emitting diodes (PHOLEDs) is one of the most important device performances and triplet hosts play a vital role of improving the QE of PHOLEDs [1]. For high QE in the PHOLEDs, good charge injection capability, good charge transport character, appropriate triplet energy and singlet energy are required [2]. In addition, thermal stability at high temperature is also necessary in order to make stable devices [3].

There are two classes of triplet host materials, and those are pure organic host materials and organometallic host materials. Organic host materials consist of various organic building blocks to construct the triplet host materials such as hole transport moieties and electron transport moieties [4,5,2]. Triplet energy, carrier transport properties and energy levels of the organic host materials can be easily managed by proper selection of the organic building blocks. Organometallic complexes are composed of a core metal and ligands coordinated to the core metal. Beryllium, zinc, and aluminum are examples of the core metals and the core metals were modified with various ligands derived from aromatic compounds. Typical ligands include 8-hydroxyquinoline [6,7], 4-hydroxybenzoquinoline [8], 10-hydroxybenzo[h]quinoline [9,10] and 2-(pyridin-2-yl)phenol [11,12]. Energy levels and charge

transport properties of the organometallic complexes are dependent on the molecular structure of the ligands and high triplet energy ligands were used to develop high triplet energy organometallic compounds. Therefore, photophysical properties of the organometallic host materials were simply manipulated by managing the ligands of the organometallic complexes [13,14]. However, not many organometallic complexes have been utilized as the host materials of PHOLEDs due to limitation of triplet energy [15,16]. In particular, only several organometallic compounds have been known as the triplet host materials for green PHOLEDs in spite of high QE of the green PHOLEDs possessing the organometallic host materials [17,18]. Therefore, it is necessary to design and synthesize more organometallic complexes through the development of new ligand structure [19]. Specifically, a thermally stable ligand structure is required because the glass transition temperature of Be compounds is not high enough and crystallization of the Be compounds can degrade the device performances at high temperature for application as high triplet energy host materials in green PHOLEDs.

In this study, we developed a Be compound, beryllium 2-(4,5-dimethyl-1-phenyl-imidazol-2-yl)phenolate (BePhIm), using a 2-(4,5-dimethyl-1-phenyl-imidazol-2-yl)phenol ligand to increase the QE of green PHOLEDs. The synthesis, material characterization and device characteristics of the BePhIm host material were studied. A high glass transition temperature of 181 °C, melting temperature of 390 °C and a high QE of 25.9% were displayed using the BePhIm host material, which is better than any other data reported using organometallic triplet host materials [12–14,19–22].

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2. Experimental section

2.1. General information

Acetic acid glacial, ethyl acetate, *n*-hexane, ethanol (Duksan Chem. Co), *o*-anisaldehyde (TCI Co), aniline, 2,3-butanedione monoxime, granular activated Zn, BeSO₄·4H₂O, and triethylamine (Aldrich Chem. Co.) were used without purification. Detailed analytical methods of synthesized compounds are described in other work [23].

2.2. Synthesis of 2-(4,5-dimethyl-1-phenyl-imidazol-2-yl)phenol

A mixture of glacial acetic acid (84 ml), *o*-anisaldehyde (7.00 g, 57.32 mmol), aniline (5.34 g, 57.32 mmol) and 2,3-butanedione monoxime (5.80 g, 57.32 mmol) was stirred in two neck round-bottomed flask under a nitrogen atmosphere for 2 h. After the mixture was cooled to room temperature, granular activated zinc (23.05 g, 352.52 mmol) was added. The mixture was stirred and refluxed for 24 h. Then, the mixture was added an ammonium hydroxide solution, and was filtered after 1 h. The filtrate was extracted with ethyl acetate and distilled water, and then the organic layer was evaporated. Crude solid was purified by column chromatography using an ethyl acetate:hexane (1:7) eluent. Yellowish white powder was obtained as a product (3.00 g, 20% yield).

¹H NMR (400 MHz, DMSO): δ 9.91 (s, 1H), 7.56 (d, *J* = 4.20 Hz, 4H), 7.28 (t, *J* = 5.80 Hz, 3H), 7.01 (t, *J* = 7.40 Hz, 2H), 2.07–1.88 (m, 6H). MS (FAB) *m/z* 264 [(M+H)⁺].

2.3. Synthesis of beryllium 2-(4,5-dimethyl-1-phenyl-imidazol-2-yl)phenolate (BePhIm)

2-(4,5-Dimethyl-1-phenyl-imidazol-2-yl)phenol (1.00 g, 3.78 mmol), BeSO₄·4H₂O (0.34 g, 1.89 mmol) and triethylamine (2.29 ml, 16.53 mmol) were dissolved in ethyl alcohol under nitrogen and the mixture was stirred at 60 °C for 3 h. The mixture was cooled to room temperature and was stirred for additional 70 h. The mixture was filtered using ethyl alcohol and was dried in vacuum oven. The final product was purified by vacuum train sublimation. White powder was obtained as a product (0.60 g, 59% yield).

¹H NMR (400 MHz, DMSO): δ 7.68 (t, *J* = 1.60 Hz, 6H), 7.50 (d, *J* = 3.40 Hz, 2H), 7.41–7.39 (m, 2H), 7.02 (t, *J* = 1.40 Hz, 2H), 6.67 (d, *J* = 3.40 Hz, 2H), 6.49 (d, *J* = 3.60 Hz, 2H), 6.13 (t, *J* = 1.60 Hz, 2H), 1.87 (s, 6H), 1.64 (s, 6H). ¹³C NMR (100 MHz, DMSO): δ 163.14, 143.29, 137.32, 133.35, 130.42, 129.85, 128.11, 127.93, 125.67, 124.88, 120.75, 116.12, 115.14, 113.02, 112.85, 9.91, 8.57. MS (FAB) *m/z* 536 [(M+H)⁺]. Anal. Calcd for C₃₄H₃₀BeN₄O₂: C, 76.24; H, 5.65; Be, 1.68; N, 10.46; O, 5.97; Found: C, 76.32; H, 5.66; N, 10.42.

2.4. Device fabrication and measurements

Green PHOLEDs were fabricated on a 50 nm thick ITO substrate at a sequence of poly(3,4-ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS, 60 nm), 4,4'-cyclohexylidenebis[N,N-bis(4-methylphenyl)aniline] (TAPC, 20 nm), N,N'-dicarbazolyl-3,5-benzene (mCP, 10 nm), BePhIm:iridium (III) tris(2-phenylpyridine) (Ir(ppy)₃) emitting layer (25 nm), diphenylphosphine oxide-4-(triphenylsilyl)phenyl (TSPO1) (30 nm), LiF (1 nm) and Al (200 nm). PEDOT:PSS was formed by spin coating and other materials were deposited by vacuum thermal evaporation. Host and dopant ratio was controlled by monitoring the deposition rate of each organic material. Doping concentration of Ir(ppy)₃ was

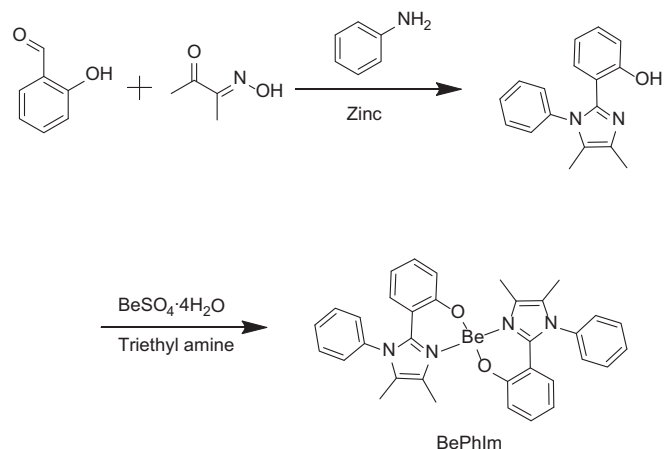
maintained as 5% and 10%. TAPC, mCP and TSPO1 were used as the hole transport layer, exciton blocking layer and electron transport layer, respectively. LiF and Al were used as electron injection layer and cathode, respectively. Electrical and optical performances of the green PHOLEDs were characterized with CS 2000 spectroradiometer and Keithley 2400 source measurement unit. QE was calculated based on an assumption of Lambertian distribution of light emission.

3. Results and discussion

The ligand synthesized in this work, 2-(4,5-dimethyl-1-phenyl-imidazol-2-yl)phenol, consists of electron rich phenol and electron poor 4,5-dimethyl-1-phenylimidazole moieties to stabilize the coordination of the ligand to Be. The main design strategy of the ligand was to realize high glass transition temperature and high triplet energy by connecting the phenol moiety with 4,5-dimethyl-1-phenylimidazole moiety, and to manage the lowest unoccupied molecular orbital (LUMO) by relatively weak electron deficiency of 4,5-dimethyl-1-phenylimidazole moiety. It was reported that the imidazole based backbone structure can raise the triplet energy of organometallic complexes because of high dipole of the molecular structure and make the LUMO level shallow [13,23]. In addition, the phenyl substituent of imidazole can improve the thermal stability of the Be compound.

The phenol and 4,5-dimethyl-1-phenylimidazole moieties were directly linked to coordinate to Be. The ligand was synthesized by common Zn catalyzed reaction between hydroxyimino ketone, aldehyde and aniline and it was coordinated to Be using hydrated BeSO₄ at a synthetic yield of 59% after purification by vacuum train sublimation [24]. ¹H and ¹³C nuclear magnetic resonance spectrometer, mass spectrometer and elemental analysis results revealed that the BePhIm host was prepared correctly. Synthetic scheme of BePhIm is shown in Scheme 1.

In order to characterize photophysical properties of BePhIm, the synthesized BePhIm host material was analyzed using ultraviolet-visible (UV-vis) spectrophotometer (UV-2501PC, Shimadzu Co.) and fluorescence spectrophotometer (F-7000, HITACHI Co.). BePhIm was dissolved in tetrahydrofuran and the concentration of sampled host material was 1.0 × 10⁻⁵ M. Phosphorescence spectrum was obtained using 2-methoxyethanol solvent at 77 K. Fig. 1 plots UV-vis absorption, solution PL and phosphorescence spectra of BePhIm. UV-vis light source was strongly absorbed at 341 nm and 354 nm by BePhIm backbone structure. The bandgap of BePhIm was measured to be 3.34 eV from UV-vis absorption edge. Solution PL emission peak of BePhIm from 2-methoxyethanol



Scheme 1. Synthetic scheme of BePhIm.

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