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Acridine derived stable host material for long lifetime blue phosphorescent organic light-emitting diodes



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ABSTRACT

A host material having acridine as a hole transport moiety, 10-(3'-(9*H*-carbazol-9-yl)-[1,1'-biphenyl]-3-yl)-9,9-dimethyl-9,10-dihydroacridine (CZBPAC), was explored as the host material of phenylimidazole type Ir triplet emitter to realize both high quantum efficiency and stable operational lifetime. The acridine containing CZBPAC was superior to carbazole based host material with the same backbone structure in that it can improve driving voltage, quantum efficiency and lifetime of the blue phosphorescent organic light-emitting diodes simultaneously.

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1. Introduction

Blue phosphorescent organic light-emitting diodes (PHOLEDs) suffer from short lifetime in spite of high external quantum efficiency (EQE) significantly surpassing that of fluorescent organic light-emitting diodes [1-4]. There are many reasons for the short lifetime of the blue PHOLEDs, but the main origin is instability of high triplet energy host materials and phosphorescent emitters. Therefore, the development of stable host and dopant materials may extend the short lifetime of the blue PHOLEDs.

In spite of the importance of the host materials for long lifetime in blue PHOLEDs, only a few host materials were reported as the stable host materials [5–12]. The most common host material for blue PHOLEDs was 3,3-di(9H-carbazol-9-yl)biphenyl (mCBP) with a biphenyl backbone structure and two carbazole functional moieties [8–12]. The mCBP host material is a hole transport type host material with a relatively shallow highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) which are matched with the HOMO and LUMO of stable Ir based phosphorescent dopant materials with a phenylimidazole ligand [4,8–16]. Generally, the phenylimidazole ligand derived Ir compounds have the HOMO level of $-5.5 \sim -5.1$ eV and the LUMO level of $-2.5 \sim -2.0$ eV. Therefore, hole transport type host materials rather than electron transport type host materials are matched with the phenylimidazole ligand based Ir emitters. Other than mCBP, hole transport type host materials such as bicarbazole derivatives [5] and 9-phenylcarbazole based dibenzothiophene derivatives performed as the host materials of the phenylimidazole based Ir emitters [7]. However, there has been no host material outperforming mCBP in terms of lifetime and a stable host material performing better than mCBP is necessary.

Herein, we report a stable host material having an acridine functional unit, 10-(3'-(9H-carbazol-9-yl)-[1,1'-biphenyl]-3-yl)-9,9-dimethyl-9,10-dihydroacridine (CZBPAC), as the host material of blue PHOLEDs. Investigation of EQE and lifetime of the CZBPAC devices demonstrated that the CZBPAC host worked better than mCBP in terms of EQE and lifetime. Voltage reduction by 1.4 V, EQE improvement by 20%, and lifetime extension by 25% were realized using the CZBPAC host instead of mCBP.

2. Experimental

General information about synthesis and analysis is described in previous work [10].





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2.1. Synthesis of 10-(3'-(9H-carbazol-9-yl)-[1,1'-biphenyl]-3-yl)-9,9-dimethyl-9,10-dihydroacridine (CZBPAC)

A solution of 9-(3'-bromo-[1,1'-biphenyl]-3-yl)-9*H*-carbazole (0.7 g, 1.7 mmol), 9,9-dimethyl-9,10-dihydroacridine (0.4 g, 1.9 mmol), potassium-*t*-butoxide (0.3 g, 2.6 mmol), tris-*tert*-butylphosphine (0.5 g, 2.6 mmol), and palladium acetate (0.1 g, 0.5 mmol) dissolved in anhydrous toluene (20 mL) was stirred in a two-necked flask under a nitrogen atmosphere followed by refluxing for 12 h. After cooling to room temperature, the solution was filtered, and filtered solution was extracted with dichloromethane and distilled water. The organic layer was dried over anhydrous MgSO₄ and concentrated. The crude product was purified by column chromatography using *n*-hexane/dichloromethane. The final product was obtained as a white powder (0.6 g, yield 62%).

¹H NMR (500 MHz, DMSO): δ (ppm) 1.63 (s, 6H), 6.24 (d, 2H, J = 4.0 Hz), 6.89 (t, 2H, J = 8.5 Hz), 6.98 (t, 2H, J = 8.5 Hz), 7.31–7.28 (m, 2H), 7.40 (d, 1H, J = 8.5 Hz), 7.43–7.44 (m, 4H), 7.49 (d, 2H, J = 9.0 Hz), 7.64 (d, 1H, J = 9.0 Hz), 7.77–7.83 (m, 3H), 7.95 (d, 2H, J = 8.0 Hz), 8.02–8.04 (m, 2H), 8.26 (d, 2H, J = 8.0 Hz). ¹³C NMR (125 MHz, CDCl₃): δ 31.3, 35.9, 109.7, 114.0, 120.0, 120.3, 120.6, 123.3, 125.3, 125.7, 126.0, 126.2, 126.3, 126.4, 126.8, 129.9, 1300, 130.4, 130.8, 131.4, 138.3, 140.7, 140.8, 141.8, 141.9, 143.0. MS (FAB) m/z 527 [(M + H)⁺]. Anal. Calcd for C₃₉H₃₀N₂: C, 88.94; H, 5.74; N, 5.32. Found: C, 88.58; H, 5.74; N, 5.29.

2.2. Device fabrication and measurement

Hole injection material and hole transport material were N.N'diphenyl-N,N'-bis-[4-(phenyl-m-tolyl-amino)-phenyl]-biphenyl-4,4'-diamine (DNTPD) and N,N,N'N'-tetra[(1,1'-biphenyl)-4-yl]-(1,1'-biphenyl)-4,4'-diamine (BPBPA), respectively. Blue triplet was tris[1-(2,4-diisopropyldibenzo[b,d]furan-3-yl)-2emitter phenylimidazole] iridium(III) (Ir(dbi)₃) and it was doped in the CZBPAC and mCBP host materials. Electron transport material was LG201. The organic materials were deposited on an indium tin oxide (ITO) substrate and a double layer electrode of LiF and Al was deposited as the cathode on the organic layers. Blue PHOLEDs for lifetime measurement had the device configuration of ITO/DNTPD (60 nm)/BPBPA (30 nm)/mCBP or CZBPAC:Ir(dbi)₃ (25 nm)/LG201 (35 nm)/LiF (1 nm)/Al (200 nm). Ir(dbi)3 was doped in the mCBP host at a doping concentration of 10%. In addition, blue devices with a stack structure of ITO/PEDOT:PSS (10 nm)/TAPC (20 nm)/mCP (10 nm)/mCBP or CZBPAC:Ir(dbi)₃ (25 nm)/TSPO1 (5 nm)/TPBI (30 nm)/LiF (1 nm)/Al (200 nm) were used to characterize basic performances. device Polv(3.4ethylenedioxythiophene):poly(styrenesulfonate) is PEDOT:PSS, 4,4'-cyclohexylidenebis[N,N-bis(4-methylphenyl)aniline] is TAPC, 1,3-bis(N-carbazolyl)benzene is mCP, diphenylphosphine oxide- 4-(triphenylsilyl)phenyl is TSPO1 and 2,2',2"-(1,3,5-benzinetriyl)tris(1-phenyl-1-H-benzimidazole) is TPBI.

Current density (J) and voltage (V) of the devices were characterized with Keithley 2400 source measurement unit and luminance (L) characteristics of the devices were collected using CS 2000 spectroradiometer. Current efficiency and power efficiency of the devices were calculated from the J, V, and L data. EQE of the devices was calculated by assuming Lambertian distribution of light emission. Lifetime measurement of the blue devices were executed at a constant current mode at an initial luminance of 500 cd/m².

3. Results and discussion

CZBPAC host material is a modified compound of mCBP with one of two carbazole units was replaced with a 9,9-dimethyl-9,10dihydroacridine (DMAC) unit. The replacement of carbazole with DMAC unit may improve hole transport property of the host material and manage the energy level to be in match with the HOMO and LUMO of Ir(dbi)₃. In particular, the HOMO level management by the DMAC unit may inject more holes and control exciton formation in the emitting layer. However, the use of two DMAC units may destabilize the host material due to poor stability of the DMAC unit for electron injection, so only one DMAC unit was introduced in the mCBP backbone structure [6,10].

Synthesis of CZBPAC was simply carried out by Pd catalyst activated coupling reaction between 9-(3'-bromo-[1,1'-biphenyl]-3-yl)-9H-carbazole and DMAC. Wet purification by column chromatography and dry purification by sublimation yielded CZBPAC as a final product. Synthesis of CZBPAC is described in Scheme 1.

The energy level characterization of CZBPAC was conducted using cyclic voltammetry (CV) measurement of ionization potential (IP). Estimated IP from the CV data in Fig. 1 was -5.75 eV. The IP value of the CZBPAC host was relatively shallower than that of mCBP (-6.00 eV) by the strong electron donating DMAC unit. As displayed in Fig. 2, the HOMO of CZBPAC was found in the DMAC unit rather than in the carbazole unit. Conversion of the IP value into the HOMO and ultraviolet–visible (UV–vis) bandgap provided the HOMO/LUMO of -5.75/-2.09 eV in the CZBPAC host. As the HOMO of CZBPAC was relatively shallow, hole injection is expected to be facilitated by adopting CZBPAC as the host material instead of mCBP.

Photophysical characterization of CZBPAC by UV–vis and photoluminescence (PL) measurements in Fig. 3 allowed evaluation of triplet energy and energy transfer of the CZBPAC host material. The triplet energy of CZBPAC was calculated to be 2.80 eV using a phosphorescent emission peak at 442 nm, which was the same as that of mCBP. Energy transfer characteristic of the CZBPAC host could be evaluated using the PL emission spectrum of CZBPAC and UV–vis absorption spectrum of Ir(dbi)₃. Large overlap of the PL emission spectrum of CZBPAC and UV–vis absorption spectrum of Ir(dbi)₃ was evident, which suggests efficient energy transfer from CZBPAC to Ir(dbi)₃.

Hole transport property of CZBPAC was compared with that of mCBP by fabricating hole only devices of CZBPAC and mCBP. Hole current density of the CZBPAC and mCBP hole only devices was presented in Fig. 4 according to driving voltage of the devices. The hole current density of the CZBPAC device was much higher than that of the mCBP device, which proposes that CZBPAC possesses better hole transport property than mCBP. It seems that the DMAC moiety facilitated hole transport through strong electron donating character.

The CZBPAC and mCBP hosts were doped with $Ir(dbi)_3$ blue dopant material at a doping concentration of 10% to compare device performances. J–V–L curves of the blue PHOLEDs are shown in Fig. 5. The J values of the blue device was high in CZBPAC device than in the mCBP device due to good hole transport and facile hole injection of CZBPAC relative to those of mCBP. The energy barrier for hole injection was 0.25 eV in the CZBPAC device, but it was 0.50 eV in the mCBP device. The L was also high in the CZBPAC device at low and high driving voltages.

Fig. 6 shows EQE-power efficiency-luminance plots of the blue PHOLEDs with the CZBPAC and mCBP host materials. Maximum EQEs of the CZBPAC device were 26.2% and 24.3%, and the EQEs at 1000 cd/m² were 25.7% and 21.8%, respectively. Both the maximum EQE and EQE at 1000 cd/m² of the CZBPAC devices were higher than those of the mCBP device. In particular, the EQE at 1000 cd/m² was dramatically enhanced in the CZBPAC device, resulting in great progress of efficiency roll-off. This behavior can be explained by the good hole injection and transport character of the CZBPAC. It has been known that the $Ir(dbi)_3$ dopant is a deep hole trap in the emitting layer due to the large HOMO level difference between host

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