

Drastic drop of external quantum efficiency at liquid nitrogen temperature in a bilayer blue phosphorescent organic light-emitting device



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ABSTRACT

We have investigated substantial drop of external quantum efficiency (EQE) at liquid nitrogen temperature in a bilayer blue phosphorescent organic light emitting device with a structure of ITO/TAPC:Flrpic/TAZ/LiF/Al, where TAPC, Flrpic, and TAZ represent 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane, iridium(III)bis[(4,6-difluorophenyl)-pyridinato-N,C^{2'}]picolate, and 3-(biphenyl-4-yl)-4-phenyl-5-(4-tert-butyl-phenyl)-1,2,4-triazole, respectively. The device exhibits a high EQE of 17.2% at room temperature although it has a simple bilayer structure. However, the quantum efficiency drops drastically to 0.8% at liquid nitrogen temperature. We studied this drastic drop of EQE in viewpoints of carrier conduction, carrier recombination, photoluminescence quantum efficiency, and energy transfer.

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1. Introduction

Since the first report on efficient bilayer organic light-emitting device (OLED) by Tang and VanSlyke [1], numerous researches have been conducted to improve the efficiency and lifetime of devices. Particularly, phosphorescent OLEDs have been widely researched in the past two decades because they can realize highly efficient displays and lighting devices by harvesting both the electro-generated singlet and triplet excitons [2]. The organic phosphors have been typically used as the guest emitters which are doped into the organic host layers inserted between functional organic layers such as carrier injection, carrier transport, and carrier blocking layers [3,4]. Utilizing these host-guest systems and multi-layer structures, the red and green phosphorescent OLEDs approaching a theoretical external quantum efficiency (EQE) of about 20% have been reported [5,6]. However, the efficient and stable blue phosphorescent device, especially having a simple organic layer structure, is still one of the important challenging issues.

Since the triplet excitons of blue phosphorescent emitters have high energies, the host materials with high triplet energies are

preferred to enhance the energy transfer and avoid the backward energy transfer from the guest to the host molecules [7–9]. In addition, the carrier transporting materials contacting with emission layer should have high triplet energies to avoid the energy transfer of triplet excitons from the emission layer to the adjacent carrier transport layers [10–12]. Therefore, the wide band gap materials with high triplet energies are typically used as host and carrier transport layers in blue phosphorescent OLEDs. These wide band gap materials may limit the carrier injection and transport so that the complicated device structures with multi-organic layers have been used to achieve a high recombination efficiency and low driving voltage. There are very few reports on simple bilayer blue phosphorescent OLEDs [13], although several red and green bilayer devices have been demonstrated [14–17].

In this paper, we report on highly efficient bilayer blue phosphorescent OLEDs. The emission layer acts as carrier injection, transport, recombination, and exciton formation in bilayer devices. Although these properties are dependent on temperature, the low temperature characteristics of the bilayer blue phosphorescent OLEDs has not been demonstrated. We observed drastic drop of external quantum efficiency at liquid nitrogen temperature in the highly efficient bilayer blue phosphorescent OLEDs. We investigated this decrease of efficiency in viewpoints of carrier conduction, carrier recombination, photoluminescence (PL) quantum efficiency, and energy transfer.

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2. Experimental

Bilayer blue phosphorescent OLEDs were prepared on the indium tin oxide (ITO) coated glass substrates. The sheet resistance of the ITO film was about $10\ \Omega/\square$. The ITO film was patterned to define anode electrodes using photolithography processes. The patterned substrates were loaded into the vacuum chamber after cleaning with isopropyl alcohol and deionized water, followed by exposing to the oxygen plasma at 10 W. Organic, LiF, and metal layers were sequentially deposited by using a vacuum thermal evaporation method in a base pressure of about 1×10^{-6} Torr. A 30 nm thick iridium(III)bis[(4,6-difluorophenyl)-pyridinato-N,C^{2'}]picolinate (Flrpic) doped 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) layer was deposited on the patterned ITO anodes. The concentration of blue phosphorescent Flrpic guest emitter was 2%. We varied the Flrpic concentration from 0.5% to 20% in the bilayer blue phosphorescent OLEDs using tris[2,4,6-trimethyl-3-(pyridine-3-yl)phenyl]borane electron transport layer [13]. The current efficiency was highest in the 2% doped device so that we fixed the Flrpic concentration to be 2% [13]. The TAPC acts as a hole transporting host material. After depositing the Flrpic doped TAPC layer, a 65 nm thick 3-(biphenyl-4-yl)-4-phenyl-5-(4-tert-butylphenyl)-1,2,4-triazole (TAZ) electron transport layer was deposited. After depositing the organic layers, a 0.5 nm thick LiF and a 100 nm thick Al layers were sequentially evaporated through a shadow mask. The completed device structure was ITO/TAPC:Flrpic (30 nm, 2%)/TAZ (65 nm)/LiF (0.5 nm)/Al (100 nm). Fig. 1 shows the chemical structures of the organic materials, the schematic device structure, and the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energy levels of the organic materials. The active area of the devices was $4 \times 4\ \text{mm}^2$. Current density-voltage-luminance characteristics of the devices were measured using computer controlled Keithley 2400 source-measure units and a calibrated fast Si photodiode (FDS010) at room temperature and liquid nitrogen temperature. The electroluminescence (EL) spectra of the devices were measured using a Minolta CS1000 spectroradiometer at room temperature and liquid nitrogen temperature.

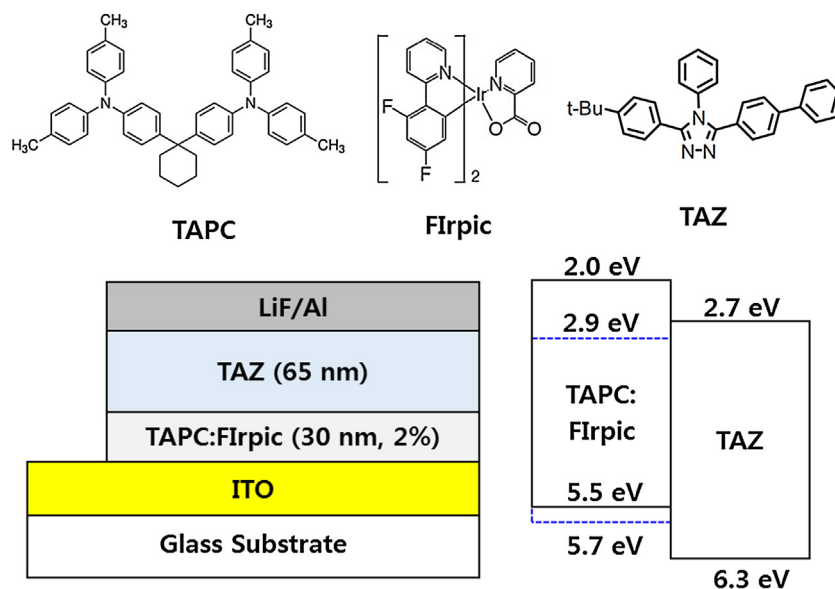


Fig. 1. Chemical structures of organic materials, device structure of the bilayer blue phosphorescent OLED, and the HOMO and LUMO energy levels of organic materials.

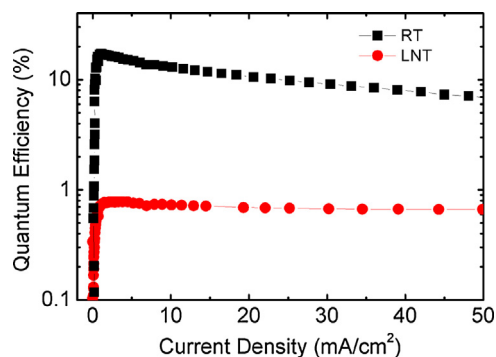


Fig. 2. EQE curves as a function of current density for the bilayer blue phosphorescent OLEDs at room (RT) and liquid nitrogen temperatures (LNT).

3. Result and discussion

Fig. 2 shows the EQE curves as a function of current density for the bilayer blue phosphorescent OLEDs measured at room and liquid nitrogen temperatures. The device structure is ITO/TAPC:Flrpic (30 nm, 2%)/TAZ (65 nm)/LiF/Al. TAPC was used as a hole transporting host for the blue phosphorescent Flrpic emitter. At room temperature, the device exhibits a maximum EQE of 17.2%, corresponding to a maximum current efficiency of 37.6 cd/A at a current density of 1.04 mA/cm². Recently, several authors reported blue phosphorescent OLEDs exhibiting maximum external quantum efficiencies of 6–11% using three organic layers [18–20]. Other authors also reported the maximum external quantum efficiencies of 10–24% using four or five organic layers [21–23]. The device shown in Fig. 1 exhibits high external quantum efficiency, although the device has a simple bilayer structure [13]. The high efficiency in our bilayer device can be attributed to the effective confinement of triplet excitons on the guest molecules and efficient recombination of injected charges [13]. Since the triplet energy of the blue phosphorescent Flrpic molecules is known to be 2.64–2.70 eV [7–9], the host material for the Flrpic should have the triplet energy higher than 2.70 eV in order to prevent the backward

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