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Recombination zone shift in phosphorescent white organic light-emitting devices with single host structure of multi-emission layers

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ABSTRACT

We have investigated the recombination zone in the phosphorescent white organic light-emitting devices with single host structure of multi-emission layers. Blue, green, and orange-red phosphorescent emitters were doped into the separate layers of single host material for fabricating the white devices with multi-emission layers. The electroluminescence spectrum was substantially modified by the shift of the recombination zone that was dependent upon the thickness of the electron transport layer. We investigated the recombination zone shift in terms of electric field distribution and carrier injection. A maximum external quantum efficiency of 15.9% and a maximum power efficiency of 28.9 lm/W were achieved by optimizing the recombination zone.

1. Introduction

Organic light-emitting devices (OLEDs) have attracted much attention in the past two decades because of their many advantages such as fast response, wide viewing angle, and thin thickness in display applications [1]. Especially, white OLEDs have been intensively researched in recent years because they can provide large area, high resolution displays and solid state lighting applications [2,3]. The external quantum efficiency of white OLEDs has been substantially improved by using phosphorescent materials which can harvest both the singlet and triplet excitons generated by the recombination of electrons and holes injected from cathode and anode, respectively [4]. The phosphorescent materials in white OLEDs have been typically used as guest emitters in the host-guest systems in order to improve the efficiency and to control the white spectrum. Various kinds of phosphorescent materials such as red, yellow, green, and blue phosphorescent emitters can be doped into the single or multiple layers to achieve the white emission [5–7]. Although the simplest one is to dope the phosphorescent emitters into a single host layer [5], it is not easy to achieve high external quantum efficiency and difficult to control the white balance. The multi-emission layers structures of single or multiple host materials have been extensively researched because of their high efficiencies and easy control of the white emission spectrum [6,7]. The white emission from phosphorescent guest molecules in the host-guest systems can be obtained by energy transfer from the host molecules to the guest emitters [8] and/or direct recombination on the guest molecules [9]. In both cases, the recombination zone acts as a critical role in achieving high efficiency and reasonable white emission spectrum.

In this paper, we demonstrate highly efficient phosphorescent white OLEDs with multi-emission layers of single host material by optimizing the recombination zone. *N*,*N*′-dicarbazolyl-3,5-benzene (mCP) was used as a common host layer. Tris(2-phenyl-1-quinoline)iridium(III) [Ir(phq)$_3$], tris(2-phenylpyridine)iridium(III) [Ir(ppy)$_3$], and iridium(III)bis[(4,6-di-fluorophenyl)-pyridinato-N,C$^{2\prime}$]picolinate (FIrpic) were used as orange-red, green, and blue phosphorescent guest materials, respectively. The guest molecules were doped into the separate layers of the mCP host. Tris[2,4,6-trimethyl-3-(pyridine-3-yl)phenyl]borane (3TPYMB) was used as an electron transport layer. We report that the emission spectrum of devices is strongly dependent on the thickness of 3TPYMB layer, which leads to substantial shift of the recombination zone. We also report the maximum external quantum efficiency (EQE) of 15.9% and power efficiency of 28.9 lm/W by optimizing the recombination zone in the white OLEDs.

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2. Experimental

Indium tin oxide (ITO) coated glass substrates were used for fabricating the phosphorescent white OLEDs. The sheet resistance of ITO film was about 10 Ω/□. After defining the ITO anode patterns using standard photolithography process, the substrates were cleaned with acetone, methanol, and deionized water. After then, oxygen plasma treatment was performed to clean the surface of ITO patterns. All organic and metal layers were deposited by using a thermal vacuum evaporation method at a base pressure of about 10^{-6}–10^{-7} Torr. A 5 nm thick 1,1-bis[(di-4-tolylamino)phenyl] cyclohexane (TAPC) layer was deposited on the patterned ITO substrate, followed by the sequential deposition of multi-emission layers of mCP:FIrpic (5 nm, 15%)/mCP:Ir(ppy)$_3$ (5 nm, 8%)/mCP:Ir(phq)$_3$ (5 nm, 10%). After then, 6.5–65 nm thick 3TPYMB layers were deposited. After depositing 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 is ITO/TAPC (5 nm)/mCP:FIrpic (5 nm, 15%)/mCP:Ir(ppy)$_3$ (5 nm, 8%)/Ir(phq)$_3$ (5 nm, 10%)/3TPYMB (6.5–65 nm)/LiF/Al. Fig. 1 shows the chemical structures of organic materials and device structure of the completed phosphorescent white OLEDs. The active area of the devices was 4 × 4 mm^2. Current density–voltage–luminance (J–V–L) characteristics of the devices were measured using computer controlled Keithley 2400 source-measure units and a calibrated Si photodiode. Electroluminescence (EL) spectrum was measured with a Minolta CS1000 spectroradiometer.

3. Results and discussion

Fig. 2 shows the EL spectra for the white OLEDs with a structure of ITO/TAPC (5 nm)/mCP:FIrpic (5 nm, 15%)/mCP:Ir(ppy)$_3$ (5 nm, 8%)/mCP:Ir(phq)$_3$ (5 nm, 10%)/3TPYMB (6.5–65 nm)/LiF/Al. The thickness of 3TPYMB layer was varied to be 6.5 nm, 35 nm, 50 nm, and 65 nm. The blue phosphorescent FIrpic molecules have two characteristic emission peaks at about 470 nm and 500 nm, which are attributed to the emissions from triplet metal to ligand charge transfer (MLCT) excited states and π–π^* ligand states, respectively [10,11]. The Ir(ppy)$_3$ and Ir(phq)$_3$ molecules have characteristic phosphorescent emission peaks at about 510 nm and 590 nm, respectively [12,13]. In the EL spectra, the device with a 6.5 nm thick 3TPYMB layer exhibits strong emission peaks at about 470 nm and 505 nm, and relatively weaker emission peak at about 575 nm. Considering that the emission peak at about 505 nm is attributed to the superimposed emissions from the π–π^* ligand states of the FIrpic molecules and triplet MLCT states of the Ir(ppy)$_3$ molecules, it can be suggested that the emission from FIrpic is stronger than the other guest molecules in the 6.5 nm thick 3TPYMB device. As the thickness of the 3TPYMB layer increases, the relative emission peak from Ir(phq)$_3$ increases, whereas the relative emissions from FIrpic and Ir(ppy)$_3$ molecules decrease. The relative emission intensities of FIrpic and Ir(ppy)$_3$ molecules become less than 0.2 in the 65 nm thick 3TPYMB device. Table 1 shows the summarized characteristics of the devices with various thicknesses of 3TPYMB layers. The increase of Ir(phq)$_3$ emission with increasing the thickness of the 3TPYMB layer results in the variation of the Commission Internationale d'Eclairage (CIE) coordinates. The 6.5 nm, 35 nm, 50 nm, and 65 nm 3TPYMB devices exhibit the CIE coordinates of (0.29, 0.44), (0.42, 0.48), (0.46, 0.46), and (0.53, 0.44), respectively, which corresponds to greenish white, greenish yellow white, yellow white, orange yellow emission colors. The *x* value in CIE coordinates substantially increases due to the increase of the Ir(phq)$_3$ emission. The emissions from guest emitters can be attributed to the energy transfer and/or direct recombination on the guest molecules. The triplet energy level of the host mCP is known to be 2.9 eV [14]. On the other hand, the guest FIrpic, Ir(ppy)$_3$, and Ir(phq)$_3$ molecules

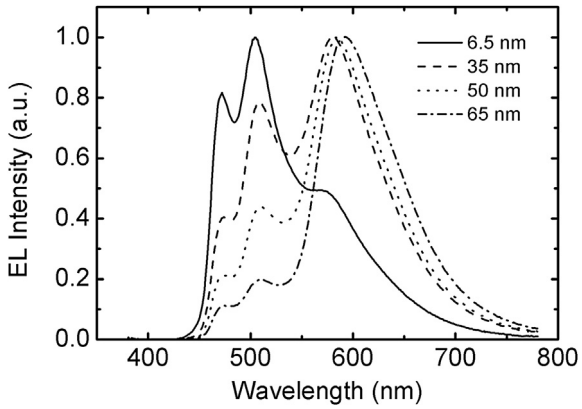

Fig. 2. EL spectra for the white OLEDs with various thicknesses of 3TPYMB layers.

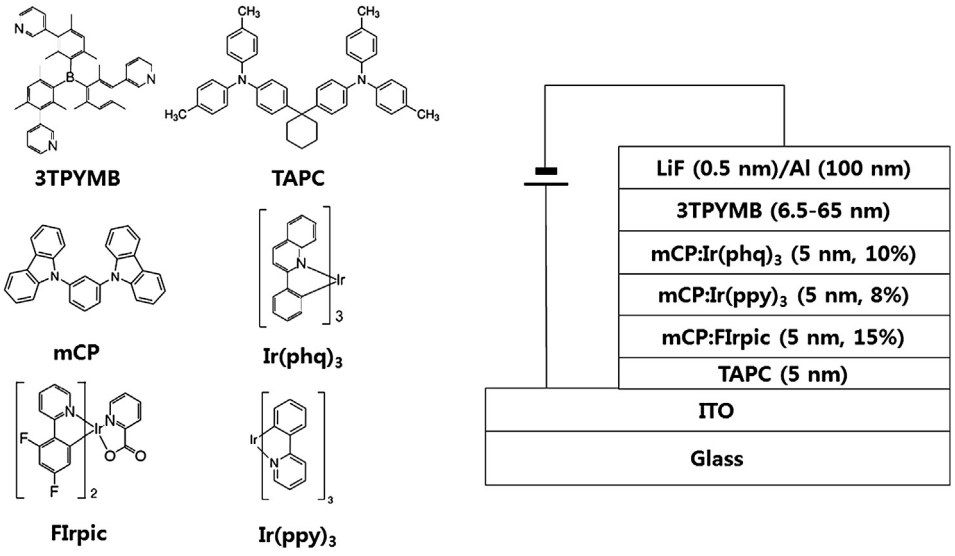

Fig. 1. Chemical structures of organic materials and device structure of the fabricated phosphorescent white OLEDs.