Relaxation of Roll-off Characteristics in Organic Electrophosphorescence Diodes

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Abstract

We demonstrate relaxation of roll-off characteristics by controlling the dopant concentrations and the thickness of an emitter layer in electro-phosphorescence diodes composed of 2,6-dicarbazolo-1,5-pyridine (PYD2)-host doped with 25 wt%-Iridium(Ⅲ)bis[(4,6-di-fluorophenyl)-pyridinato-N,C^2] picolinate (FIrpic).

1. Introduction

Organic light emitting diodes (OLEDs) have been attracted much attention as the next generation flat panel display. Recently, by introducing the phosphorescence materials such as Ir complexes, high external electroluminescence efficiencies have been achieved [1][2]. However, phosphorescence OLEDs showed the significant roll-off characteristics under a high current density due to the triplet-triplet annihilation [3][4]. Such characteristics become a serious problem in terms of passive matrix operation which requires high current density near J ～ 1 A/cm^2.

In this study, we report relaxation of roll-off characteristics by controlling the concentrations of guest molecules and the thickness of the emitting layer in electrophosphorescence devices.

2. Experimental

Absolute PL quantum efficiency (Φ_{PL}) was measured by an integrating sphere (Is-060, Labsphere Co.) [6]. Absorption and photoluminescence (PL) spectra of organic deposited films were measured by

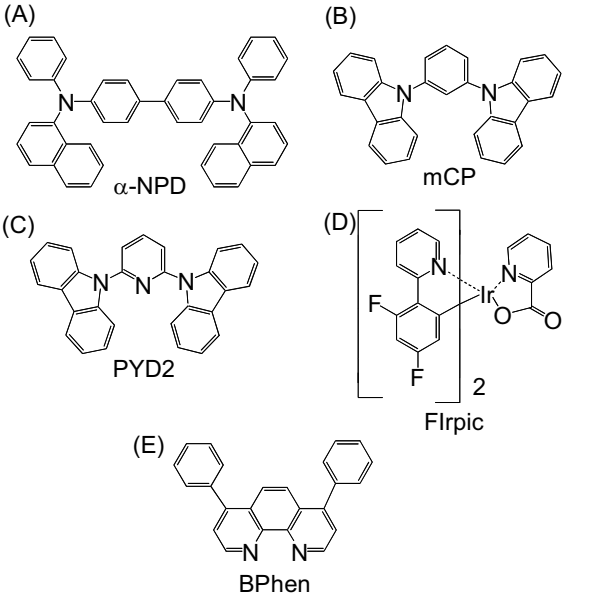

Fig. 1. Chemical structures of hole injection layer [(A) :α-NPD], hole transport layer [(B) :mCP], host [(B) :PYD2], guest [(D) :FIrpic] materials and electron transport layer [(E) :BPhen] used in this study.

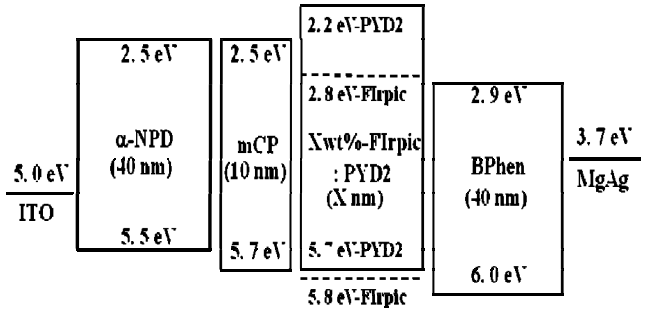

Fig. 2. OLED structure used in this study. HOMO, LUMO levels of hole injection layer [α-NPD], hole transport layer [mCP], emitting layer [PYD2, FIrpic] and electron transport layer [BPhen] with work functions of indium thin oxides [ITO] and MgAg electrode are indicated.

UV-2550 (SHIMADZU Co.) and FP-6500 (JASCO Co.) spectrometers, respectively. Transient fluorescence and phosphorescence spectra were measured by a streak camera (Hamamatsu C4334). The highest occupied molecular orbital (HOMO) levels of organic layers were measured with an ultraviolet photoelectron spectroscope (Riken Keiki, AC-1) and the lowest unoccupied molecular orbital (LUMO) levels were calculated from subtraction of the energy gap (E_g) values from their HOMO levels. 50 nm-thick organic films were prepared by high-vacuum (1.0×10^{-3} Pa) thermal evaporation onto pre-cleaned silicon and quartz substrates. For these measurements, OLEDs were fabricated according to the following steps. Indium tin oxide (ITO) coated glass substrates were degreased with solvents and cleaned in a UV-ozone chamber for 20 min. Then, organic layers and metal electrodes were deposited by high-vacuum (1.0×10^{-3} Pa) thermal evaporation onto pre-cleaned ITO coated glass substrates. Firstly, 40nm-thick-4,4'-bis[N(1-naphthyl)-N-amino]biphenyl (α-NPD) as a hole injection layer (HIL) was deposited. Second, 10 nm-thick-N,N'-dicarbazolyl-3, 5-benzene (mCP) as a hole transport layer (HTL) was deposited. As an emitting layer (EML), 25 wt%-Iridium(Ⅲ) bis[(4,6-di-fluorophenyl)-pyridinato-N,C^2] picolinate (FIrpic) doped 2,6-dicarbazolo-1,5-pyridine (PYD2) was deposited. Finally, a 40 nm-thick 4,7-diphenyl-1,10-phenanthroline (BPhen) as an electron transport layer (ETL) was deposited. A MgAg layer with an atomic ratio of 10:1 as the cathode and a 10 nm-thick Ag capping layer were deposited through a 1 mm-diameter opening in a shadow mask. J-V characteristics were measured using a semiconductor parameter analyzer (E5250A, Agilent Technologies) and EL spectra were obtained using a multichannel spectrometer (Ocean Optics, UBS2000).

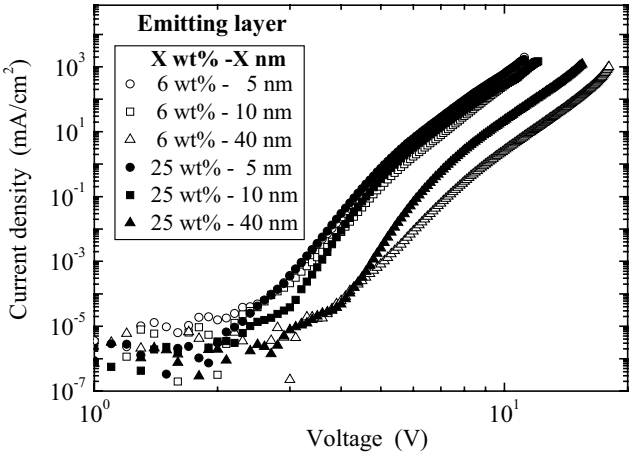

Fig. 3. Current density and voltage (J-V) characteristics dependence on FIrpic concentration and thickness emitting layer in ITO (160 nm) / α-NPD (40nm) / mCP (10 nm) / X wt%-Firpic in PYD2 (X nm) / BPhen (40 nm) / MgAg (100 nm) / Ag (10 nm).

3. Results and discussion

Figure 1 shows the chemical structure of the α-NPD, mCP, PYD2, FIrpic, and BPhen used in this study. PYD2 has a high triplet energy level,

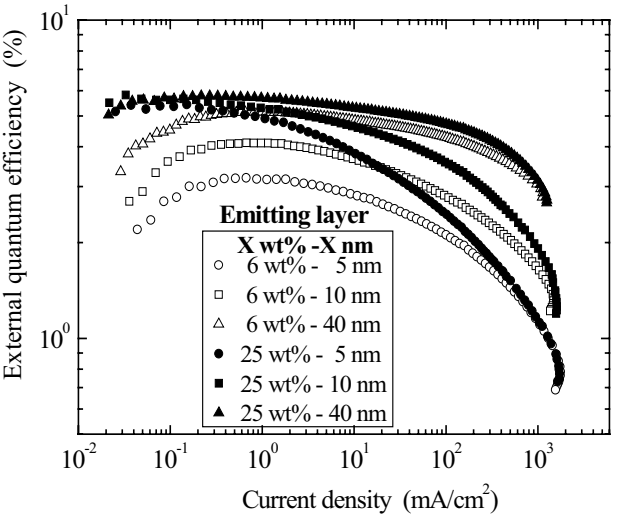

Fig. 4. External quantum efficiency and current density (η_{EL}-J) characteristics dependence on FIrpic concentration and thickness emitting layer in ITO (160 nm) / α-NPD (40nm) / mCP (10 nm) / X wt%-FIrpic in PYD2 (X nm) / BPhen (40 nm) / MgAg (100 nm) / Ag (10 nm).

providing excellent host in blue phosphorescence OLEDs [6].

Figure 2 shows the OLED structure with their HOMO and LUMO levels including the work functions of the ITO and MgAg layers. Here, we mention that the electron injection barrier between BPhen and PYD2 interface is 0.7 eV which is significantly higher than that between BPhen and FIrpic energy barrier (0.1 eV).

Figure 3 shows the current density (J) - voltage (V) characteristics depending on the FIrpic concentration and the thickness of their EML. The J-V curves shifted to the lower driving voltage as an increase of the doping concentration. This phenomenon is due to the enhancement of electron injection efficiency by increasing the FIrpic concentration [6].

Figure 4 shows η_{EL} depending on the thickness of EML and the FIrpic concentrations. In case of 25 wt%-FIrpic doped devices, they showed almost same η_{EL} at the low current density regions of 0.01 mA/cm^2 ~ 1 mA/cm^2. However, at the high current density region of 1 mA/cm^2 ~ 1000 mA/cm^2, significant differences in η_{EL} was observed depending on the EML thickness. The narrower EML thickness resulted in quicker roll-off characteristics. In case of the 6 wt%-FIrpic doped devices, the similar roll-off characteristics were observed.

Figure 5 (a) shows EL spectra depending on the FIrpic concentrations and the EML thicknesses, and Fig. 5 (b) shows the short wavelength area (350 nm ~500 nm) of the EL spectra. 25 wt%-FIrpic doped devices were consisted of only FIrpic PL component, although the 6 wt%-FIrpic doped devices showed the additional PL components of α-NPD, BPhen and FIrpic. Further, the non-doped device showed the EL components of only α-NPD and BPhen, and no PL component of PYD2 was observed, indicating that non-doped PYD2 layer does not provide efficient carrier recombination site. In the FIrpic doped devices, on the other hand, electrons are directly trapped by the FIrpic LUMO level based on the energy level, leading to direct exciton formation at FIrpic. Therefore, in case of low concentration (6 wt%) devices with thin EML thickness (5 nm and 10 nm) sufficient carrier trapping has not been realized, leading to low η_{EL}. Further, we observed that the roll-off characteristics are appreciably relaxed in the devices with thicker EML. With a 40 nm-thick emitter layer, an η_{EL} of ~3% was maintained even at J= 1 A/cm^2 that would be explained by the lower exciton density in EML. Thus, the relaxation of the roll-off characteristics can be obtained by controlling the EML thickness and the guest concentrations by taking into account of electron injection and transport characteristics of FIrpic.

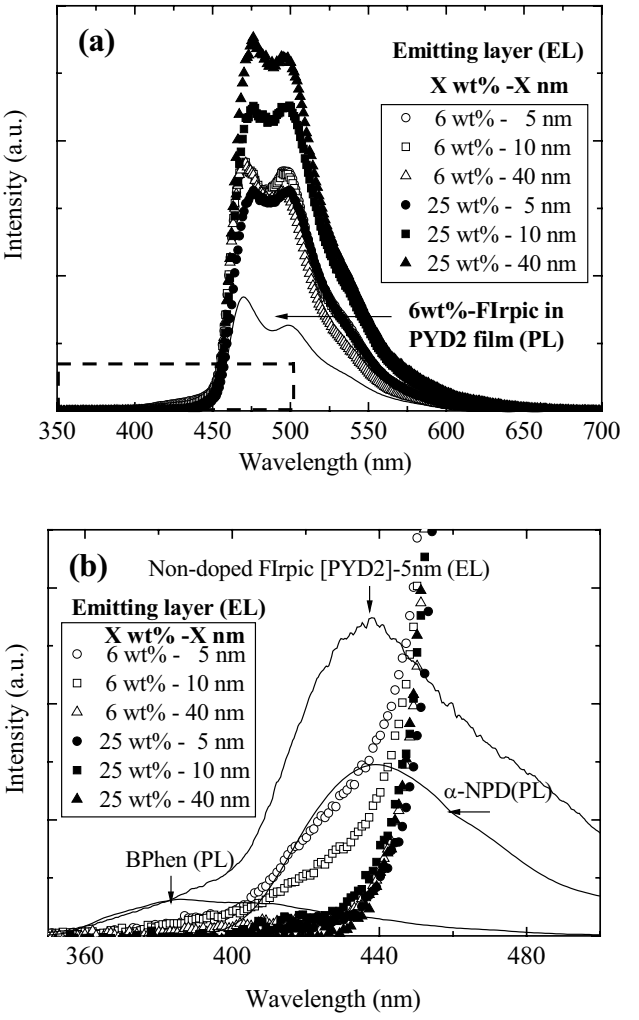

Fig. 5. EL spectra in ITO (160 nm) / α-NPD (40nm) / mCP (10 nm) / X wt%-FIrpic in PYD2 (X nm) / Bphen (40 nm) / MgAg (100 nm) / Ag (10 nm) at J = 10 mA/cm^2 depending on various FIrpic concentration and thickness and PL spectrum of 6wt%-FIrpic in PYD2 film (50 nm) (a), and (b) shows expanded short wavelength[dotted square] part of [A] including BPhen and α-NPD neat film.

4. Summary

Controlling the FIrpic concentration was found to improve η_{EL} by enhancing electron current into the EML. Also, roll-off characteristics of η_{EL} were greatly improved by controlling the emitter thickness, which suppresses triplet-triplet exciton annihilation at high current density.

5. References

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