White Organic Light-emitting Diodes using the Tandem Structure Incorporating with Organic p/n Junction

Hyun Koo Lee**, Do Sung Kwon**, and Chunghee Lee*

Abstract

Efficient white organic light-emitting diodes are fabricated with the blue and red electroluminescent (EL) units electrically connected in a stacked tandem structure by using a transparent doped organic p/n junction. The blue and red EL units consist of the light-emitting layer of 1,4-bis(2,2-diphenyl vinyl)benzene (DPVBi) and 4-dicyanomethylene-2-methyl-6-[2-(2,3,6,7-tetrahydro-1H,5H-benzo[i,j] quinolizin-8-yl)vinyl]-4H-pyran) (DCM2) doped *tris*(8-hydroxyquinoline) aluminum (Alq_3), respectively. The organic p-n junction consists of α-NPD doped with $FeCl_3$ (15 % by weight ratio) and Alq_3 doped with Li (10 %). The EL spectra exhibit two peaks at 448 and 606 nm, resulting in white light-emission with the Commission Internationale d'Eclairage (CIE) chromaticity coordinates of (0.36, 0.24). The tandem device shows the quantum efficiency of about 2.2 % at a luminance of 100 cd/m^2, higher than individual blue and red EL devices.

Keywords : White organic light-emitting diode (OLED), tandem structure, organic p/n junction

1. Introduction

Generation of white light from organic light-emitting diodes (OLEDs) has attracted great attention due to their applications in full-color displays, backlights, and solid-state lightings.[1] Full-color method using the combination of efficient white OLEDs and color filters has is advantages such as it does not require high resolution of the shadow mask for separately depositing red, green and blue pixels. Furthermore, efficient white OLEDs can replace conventional incandescent and fluorescent lamps for various lighting applications such as a backlight for LCDs.

Various device structures such as the multilayer structure of several emitting layers, doping several organic molecules with different colors in a host emitting layer, or blending several light-emitting polymers have been utilized to generate white electroluminescence (EL).[1-8] In these device structures, careful control of doping concentration, blending ratios and/or the thickness of each emitting layer is necessary to obtain a balanced white light emission by preventing a cascade energy transfer from blue to green and red. In addition, the EL spectra and the chromaticity coordinates of such devices can change with the bias voltage or current density due to the movement of electron-hole recombination region and/or the change of the concentration of excitons over several emitting layers. Therefore, it is a great challenge to develop high efficiency white OLEDs with stable color coordinates.

Recently, L. S. Liao *et al.*[9] reported high-efficiency tandem organic light-emitting diodes, with multiple EL units connected electrically in series by using an optically transparent doped organic p/n junction. The tandem structure has the advantage in that the emission light from each EL unit is practically independent, as if individual EL devices are connected in series.[9,10] This property offers great freedom for fabricating white light-emitting diodes since one can easily tune the relative intensity of emission lights from individual EL units with different colors in a tandem structure without the problem of the cascade energy transfer or the chromaticity changes with the bias voltage, commonly observed in devices with multiple emitting layers.

In this paper, we report the fabrication and characterization of tandem white OLEDs consisting of blue and

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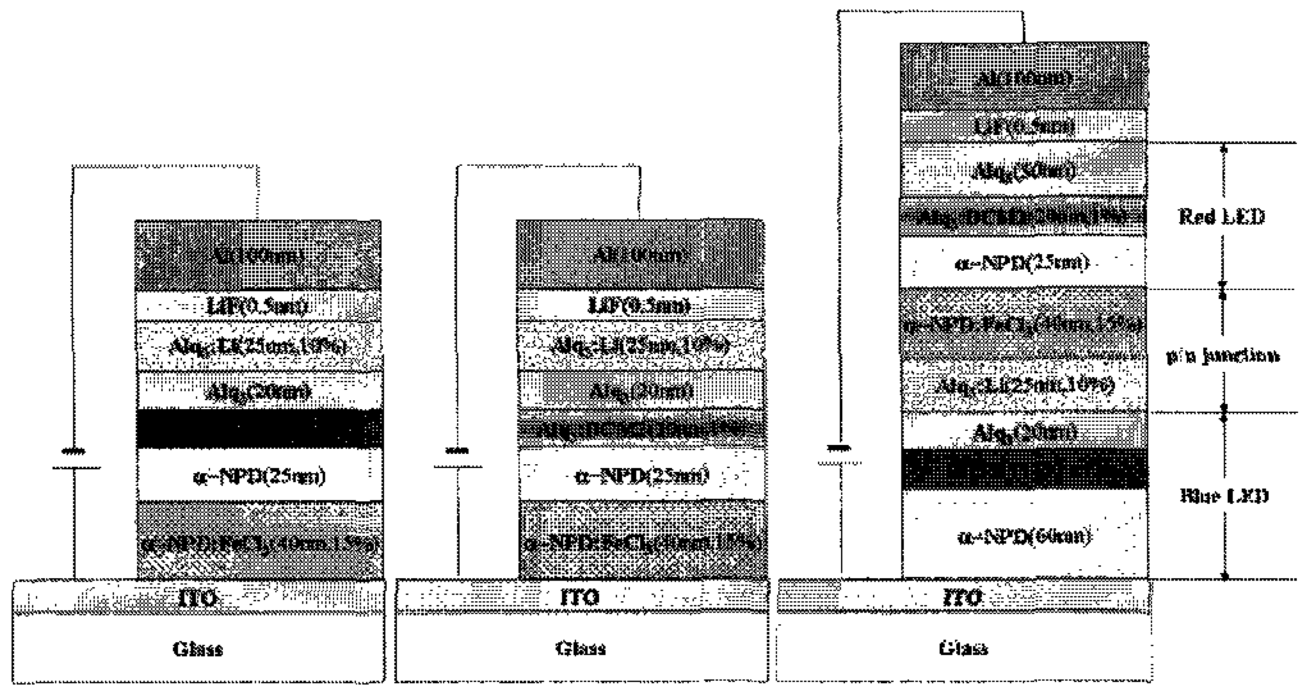
*Member, KIDS ; **student Member, kids
Corresponding Author : Changhee Lee
School of Electrical Engineering and Computer Science and Inter-University Semiconductor Research Center, Seoul National University, Seoul 151-744, Korea
E-mail : chlee7@snu.ac.kr **Tel** : 02-880-9093 **Fax** : 02-877-6668

red EL units vertically connected with the doped organic p/n junction. The blue and red EL units consist of the light-emitting layer of 1,4-bis(2,2-diphenyl vinyl)benzene (DPVBi) and 4-dicyanomethylene-2-methyl-6-[2-(2,3,6,7-tetrahydro-1H,5H-benzo[i,j] quinolizin-8-yl)vinyl]-4H-pyran) (DCM2) doped *tris*(8-hydroxyquinoline) aluminum (Alq_3), respectively. The device shows a white light emission with an external quantum efficiency (QE) of 2.2 % and the CIE coordinates of (0.36, 0.24).

2. Experimental

Organic light-emitting devices were fabricated by successive vacuum deposition of individual EL unit, organic p/n junction and LiF/Al cathodes under a vacuum of about $2x10^{-6}$ Torr on the ITO substrates. The ITO substrates were cleaned with sequential ultrasonication in organic solvents such as isopropyl alcohol, acetone and methanol, and rinsing in a de-ionized water and then treated with UV ozone. Fig. 1 shows the device structure of individual EL units and the tandem white OLEDs consisting of blue and red EL units. The individual EL units consist of a hole-transporting layer of 4,4'bis[N-(1-napthyl)-N-phenyl-amino]-biphenyl (α-NPD), light-emitting layer, and an electron-transporting layer of Alq_3. The light-emitting layer of the blue and red EL unit is DPVBi and DCM2 doped Alq_3 (doping concentration of 1 %), respectively. The transparent organic p-n junction consists of the p-type doped layer of α-NPD doped with $FeCl_3$ (15 % by weight ratio) and the n-type doped layer of Alq_3 doped with Li (10 %). The thickness of each layer, the concentration of DCM2, and the concentration of Li and $FeCl_3$ were tuned to obtain a white light emission. After depositing organic materials, the Al cathode with a very thin (1 nm) layer of LiF was deposited without breaking vacuum. The evaporation rate was 1~2 Å/sec for organic materials and 4~5 Å/sec for Al deposition. The overlapping area of the ITO and the Al cathode was 1.96 mm^2.

The devices mounted in the sample holder in vacuum and all measurement was carried out at room temperature. The current-voltage (I-V) characteristics were measured with a Keithley 236 source-measure unit. The intensity of the EL emission was simultaneously measured with a Keithley 2000 multimeter equipped with a calibrated Si photodiode (Hamamatsu S5227-1010BQ) or a photomultiplier tube (ARC P2 PMT) through an ARC 275 monochromator.



(a) (b) (c)

Fig. 1. The schematic diagram of the device structures for (a) blue EL device, (b) red EL device and (c) a tandem white OLED consisting of blue and red EL units electrically connected by an organic p/n junction.

3. Results and Discussion

Fig. 2 exhibits the current-voltage-luminance (I-V-L) characteristics for the blue and red EL devices of the structure shown in Figs. 1 (a) and (b), respectively. The inset shows the external quantum efficiency of the devices as a function of the current density. The red EL device shows a lower EL onset voltage (~3 V) and higher QE (~ 1.6 %) compared with the blue EL device which shows the EL onset voltage of 4 V and the QE of ~ 0.9 %. The QE of the red EL device decreases with the current density due to the exciton quenching effect.[11]

Fig. 3 compares the I-V-L characteristics of two tandem OLEDs having different order of the red and blue EL units, ITO/Blue/n-p/Red/Al and ITO/Red/n-p/Blue/Al. The inset compares the quantum efficiency of the two devices as a function of the current density. Two devices show similar I-V characteristics with the same EL onset voltage of about 7 V. This EL onset voltage is the sum of two unit devices, 3 V for the red EL unit and 4 V for the blue EL unit, as shown in Fig. 2. This behaviour can be understood as in a voltage divider since the bias voltage applied to the tandem device is divided by the impedance ratio of each unit device. Because the red EL device shows higher current at a given voltage compared with the blue EL device, as shown in Fig. 2,

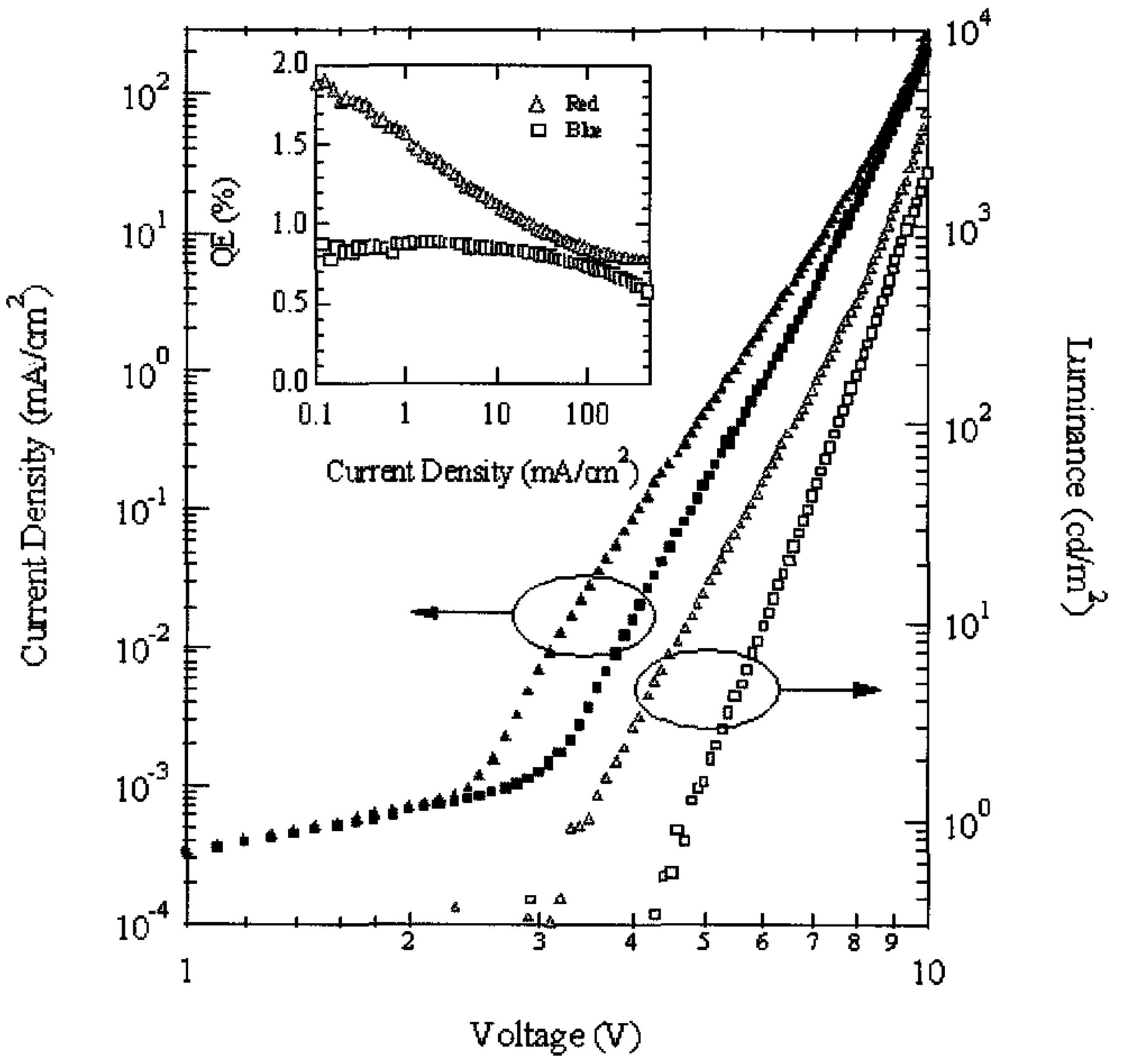


Fig. 2. Current-voltage-luminance characteristics of the blue (square) and red (triangle) OLEDs shown in Figs. 1 (a) and (b), respectively, measured at room temperature. The inset shows the quantum efficiency of the devices as a function of the current density.

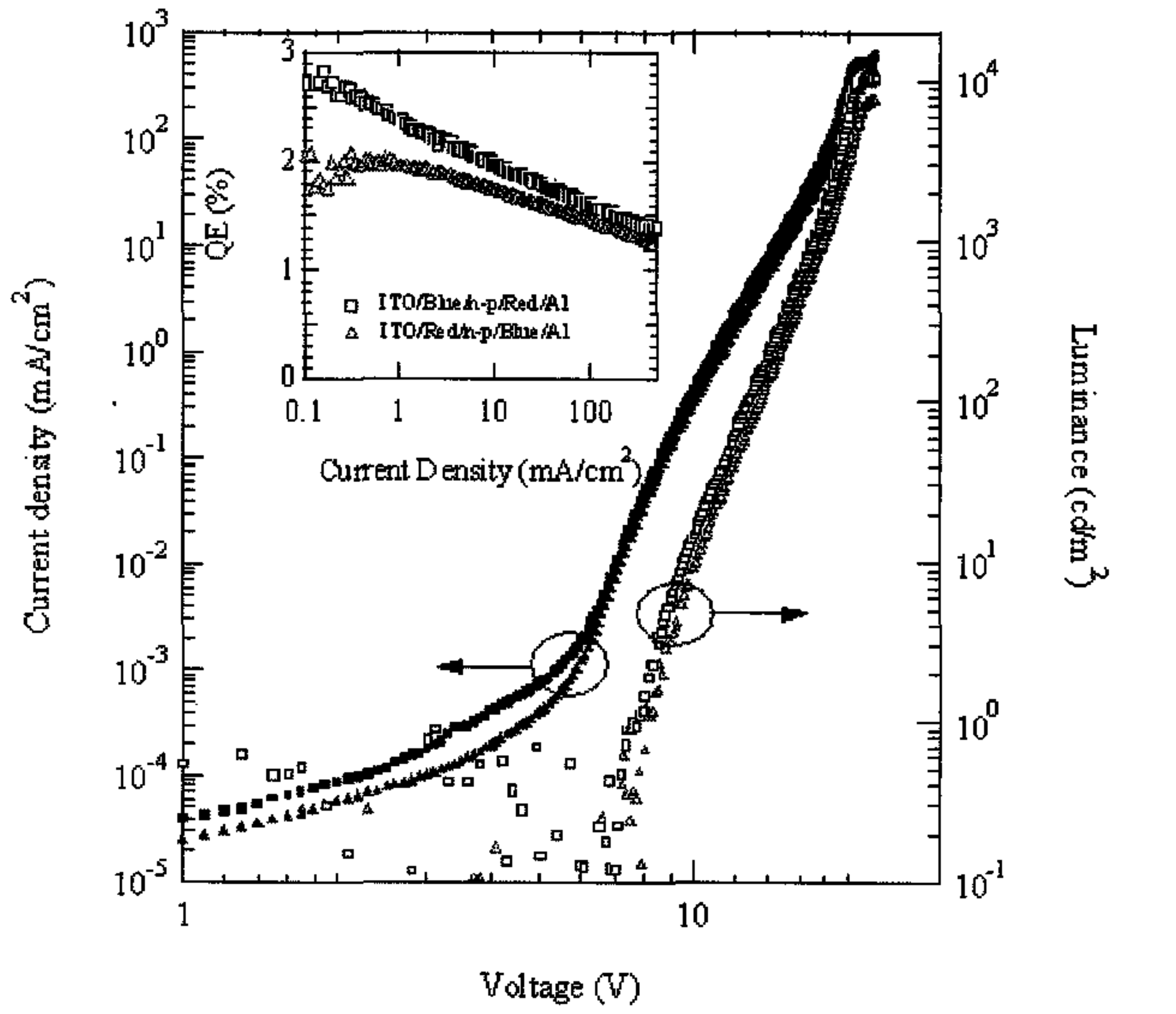


Fig. 3. Comparison of the I-V-L characteristics of two types of tandem OLEDs with the device structure of ITO/Blue/n-p/Red/Al (square) and ITO/Red/n-p/Blue/Al (triangle) OLEDs, measured at room temperature. The inset compares the quantum efficiency of the devices as a function of the current density.

the effective conductance of the red one is higher than the blue one. Therefore, the I-V dependence of the tandem device is mainly determined by the blue unit since the larger potential difference is applied to the blue unit. Therefore, the I-V dependence is similar for two tandem devices and the EL onset voltage is the sum of the red and blue EL units.

The QE is slightly higher for the device structure of ITO/Blue/n-p/Red/Al compared with ITO/Red/n-p/Blue/Al, ever although the two devices show similar I-V characteristics. This QE difference implies the electron-hole balance in each EL unit changes depending on the stacking order of two units. To clarify this QE difference, it is necessary to investigate the relative efficiency of the electron and hole recombination in each EL unit.

Fig. 4 compares the EL spectra of two types of tandem devices under a current density of 26 mA/cm^2. The EL spectra is the sum of the EL spectra of two individual EL devices, consisting of two peaks around 450 nm emitted from DPVBi and 606 nm emitted from DCM2. Although the I-V characteristics are similar for both device structures, as shown in Fig. 3, the relative intensity of two EL peaks are different for the two types of tandem devices. The device with the structure of ITO/Red/n-p/Blue/Al shows predominantly blue EL emission and its red EL emission is very weak. However, both red and blue EL peaks are comparable for the devices with the ITO/ Blue/ n-p/Red/Al structure. The result indicates that the

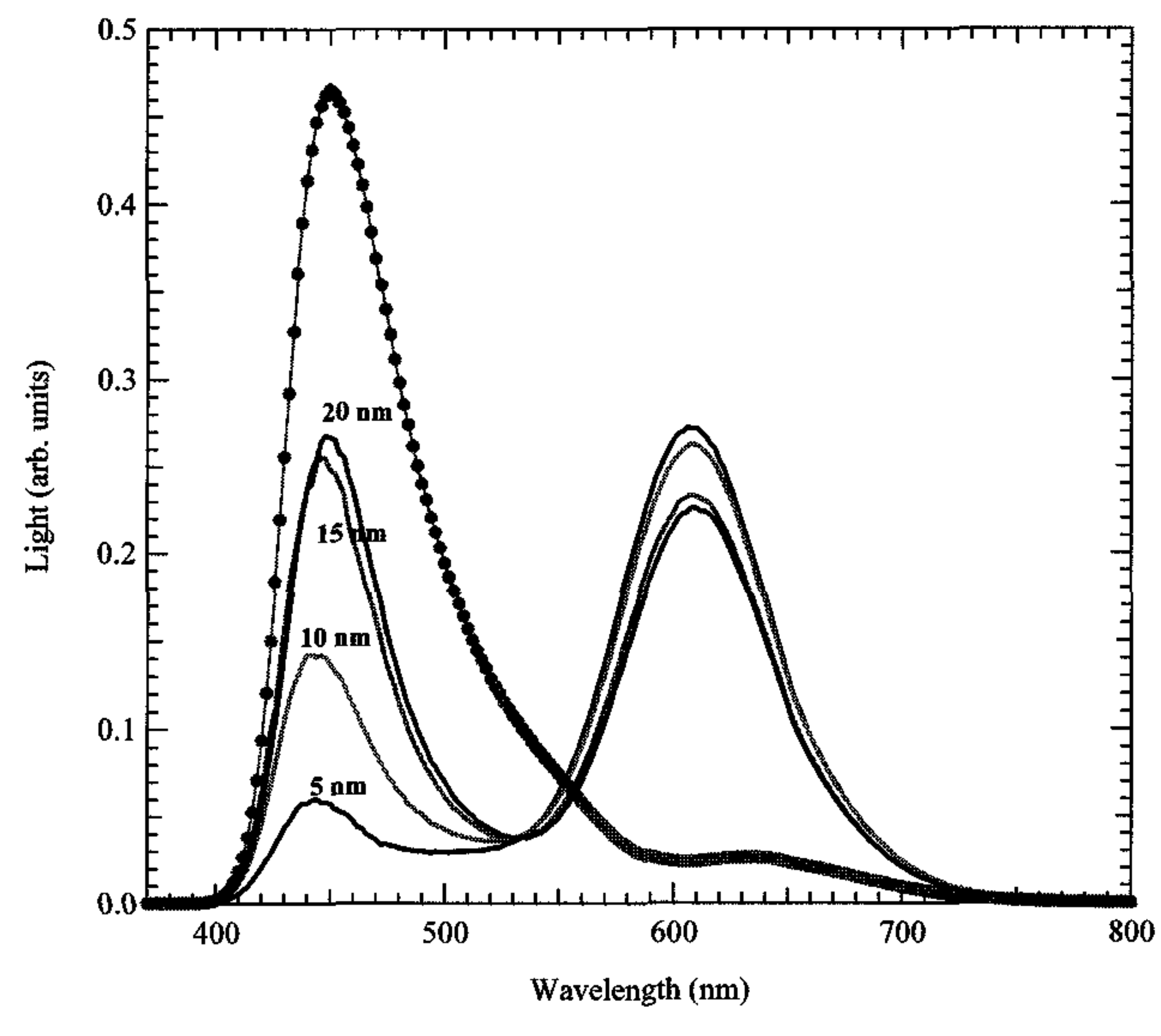


Fig. 4. Comparison of the EL spectra of two types of tandem devices under a current density of 26 mA/cm^2. For the device structure of ITO/Blue/n-p/Red/Al, the thickness of the DPVBi layer in the blue EL unit was changed from 5 to 20 nm as indicated in the figure.

electron- hole (e-h) recombination occurs efficiently in the blue EL unit irrespective to the stacking order. However, the e-h recombination is not efficient for the case where the red EL unit is located in the first stack on the ITO substrates compared with the case where the red unit is located in the second stack. This implies that the charge generation at the organic p/n junction is not very efficient. If the electron injection from the organic p/n junction into the first stack is limited, the e-h balance in the first EL stack worsen, resulting in poor EL efficiency for the red EL unit in the first stack. Although the e-h balance is not efficient in the first EL stack, the blue EL emission can occur comparably for the case where the blue EL unit is located in the first stack since a large portion of the bias voltage is divided into the blue EL unit. Then, the electron injection from the organic p/n junction into the first stack can be enhanced at high field.

Since the device structure of ITO/Blue/n-p/Red/Al provides comparable light emission from both EL units, we chose this device structure to fabricate the white tandem OLEDs. The thickness of the DPVBi layer in the blue EL unit was changed from 5 to 20 nm, while other layers were not changed, in order to control the relative intensity of the blue peak for obtaining white light emission. The blue emission increases as the thickness of the DPVBi layer increase, while the red emission intensity decreases only slightly. This behavior clearly indicates that each EL unit can be independently controlled for the tandem device structure. Fig. 4 shows that white light emission is generated for the DPVBi thickness of 15-20 nm.

Fig. 5 compares the EL spectra normalized at the peak wavelength for the tandem OLED with the DPVBi thickness of 20 nm and individual blue and red devices under a current density of 26 mA/cm2. Clearly, the EL spectra of the tandem structure are a sum of the EL spectrum of two individual devices, resulting in white emission with the CIE chromaticity coordinates of (0.36, 0.24). Interestingly, the blue emission peak is slightly shifted to the shorter wavelength and the spectral width is narrower for the tandem structure compared with the blue EL unit device. This behaviour can be attributed to the optical microcavity effect since the blue emission zone of the tandem structure is farther apart from the reflective metal cathode compared with the blue unit device.

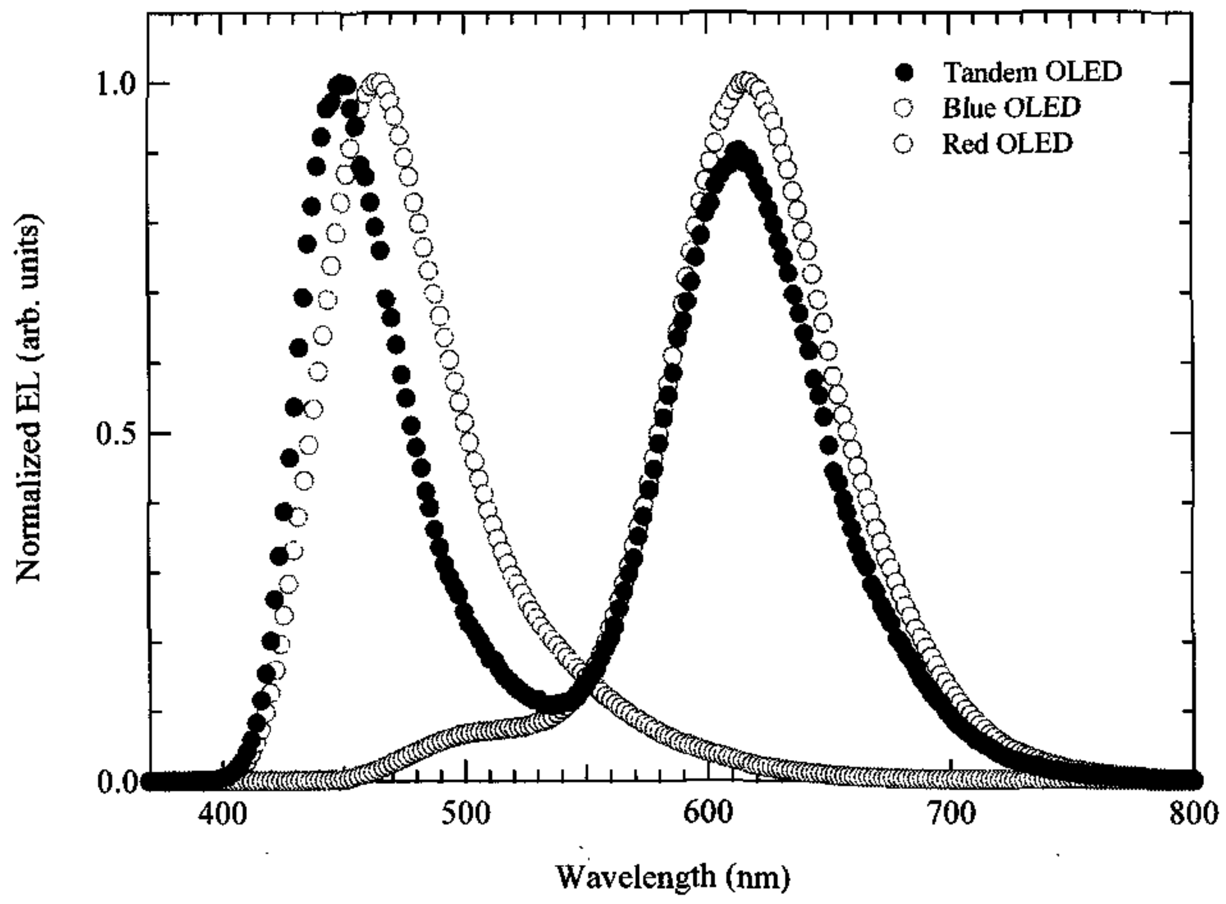


Fig. 5. EL spectra normalized at the peak wavelength for the tandem OLED with the DPVBi thickness of 20 nm and individual blue and red devices under a current density of 26 mA/cm^2.

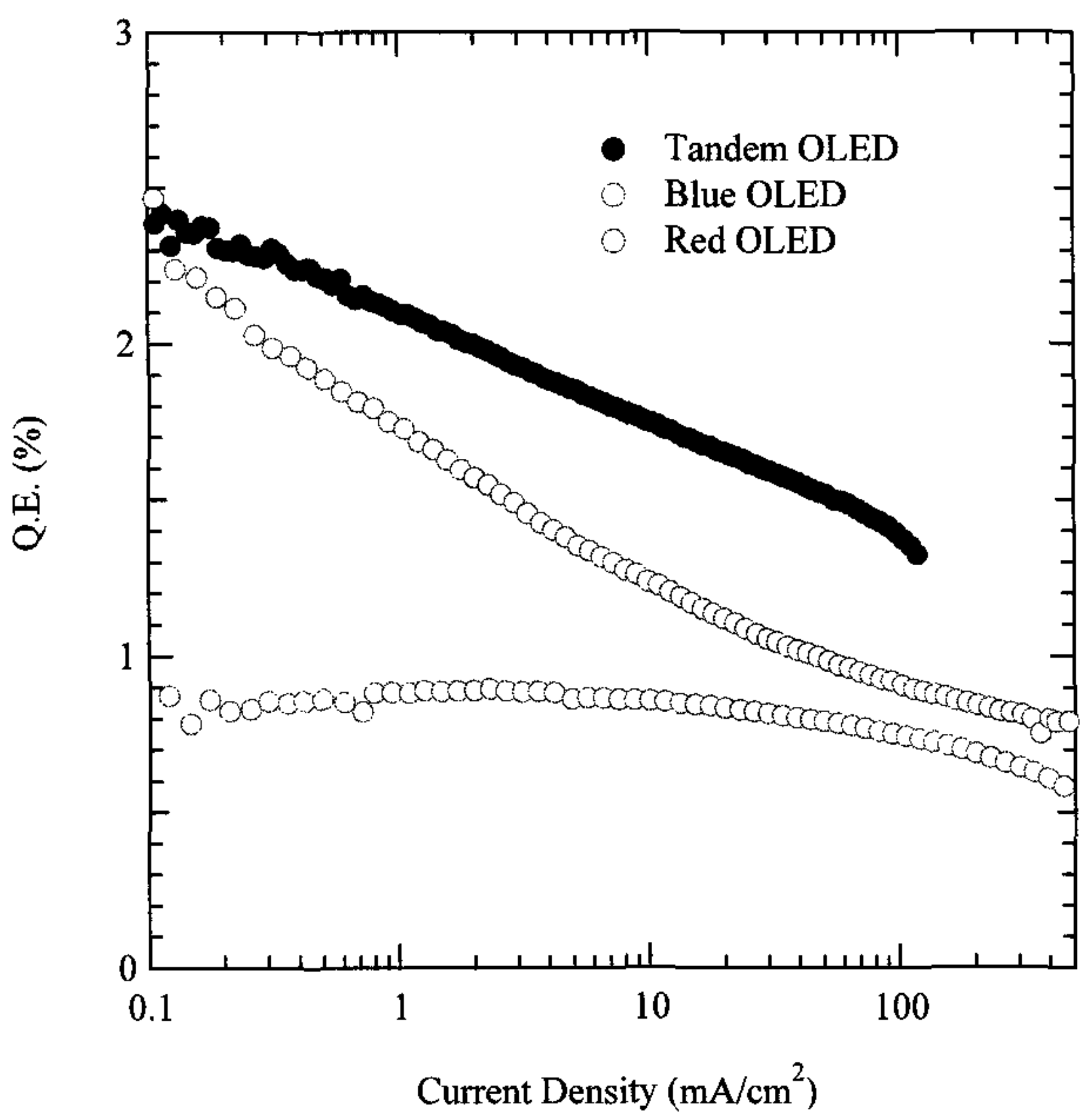


Fig. 6. Comparison of external quantum efficiency of the tandem OLED with the DPVBi thickness of 20 nm and individual blue and red devices as a function of the current density.

Fig. 6 compares the external quantum efficiency of the tandem OLED with the DPVBi thickness of 20 nm and individual blue and red devices as a function of the current density. Since the light from two EL units is added with the same current flowing through them, the quantum efficiency is higher for the tandem device compared with the individual blue and red devices. The tandem device shows the quantum efficiency of about 2.2 % at a luminance of 100 cd/m^2.

4. Conclusions

This work demonstrated that efficient white tandem OLEDs can be fabricated with the blue and red EL units electrically connected by using a transparent organic p/n junction. The blue and red EL units consist of the light-emitting layer of DPVBi and DCM2-doped Alq_3, respectively. The ITO/Blue/n-p/Red/Al structure shows higher QE and comparable red and blue light emission compared with the ITO/Red/n-p/Blue/Al structure. We believe that such difference occurring according to the stacking order of the blue and red unit is attributed to the combined effect of the impedance difference of two unit devices and poor charge generation efficiency of the organic p/n junction. The EL spectra of the ITO/Blue/n-p/Red/Al structure are a sum of two individual EL spectra. The thickness of the DPVBi layer in the blue EL unit can be controlled to obtain a white light emission without affecting the EL characteristics of the red EL unit. White light emission with the CIE chromaticity coordinates of (0.36, 0.24) is obtained for the DPVBi thickness of 20 nm. Since the light from two EL units is added with the same current flowing through them, the QE is higher for the tandem device (~2.2 % at a luminance of 100 cd/m^2) compared with the individual blue and red devices.

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