

Efficient White Organic Electroluminescent Devices Consisting of Two Emitting Layers of Blue and Orange Colors

Namheon Lee, Munjae Lee, Jun-Ho Song, Changhee Lee and Soon-Ki Kwon¹

Department of Physics, Inha University, Incheon 402-751, Korea

¹Department of Polymer Science & Engineering and Research Institute of Industrial Technology, Gyeongsang National University, Chinju, 660-701, Korea

Phone: +82-32-860-7666, E-mail: chlee7@inha.ac.kr

Abstract

We report efficient white organic electroluminescent devices consisting of a blue-emitting layer of 9,10-bis[(2'',7''-di-*t*-butyl)-9',9''-spirobifluorenyl]anthracene (TBSA) and a red-emitting layer of 4-dicyanomethylene-2-methyl-6-[2-(2,3,6,7-tetrahydro-1*H*,5*H*-benzo[*i,j*]quinolizin-8-yl)vinyl]-4*H*-pyran) (DCM2) doped into 4,4'-bis[*N*-(1-naphthyl)-*N*-phenyl-amino]-biphenyl (α -NPD). The device shows the CIE coordinates of (0.32, 0.37). The external quantum efficiency is about 3.4 % and the luminous efficiency is about 3.9 lm/W at luminance of 100 cd/m². The maximum luminance is about 45,400 cd/m² at 11.5 V.

1. Introduction

Organic light-emitting diodes (OLEDs) have been extensively studied as a new type of flat-panel display since C. W. Tang and S. A. Van Slyke reported highly efficient OLEDs using Alq₃ [1]. In general, full-color OLEDs are fabricated by patterning RGB (red, green, blue) subpixels using vacuum deposition with a shadow mask or inkjet-printing. Other full-color methods such as combining white emission with a color filter or the color conversion method in which the blue emission was converted to green and red are also promising. The combination of a white OLED and a color filter has advantages since it does not require very accurate alignment of the shadow mask and it has less deposition steps for organic emitting layers. However, there is a sacrifice in the luminous efficiency due to the absorption of emission light by a color filter. Therefore, it is very important to develop white OLEDs with high brightness and high luminous efficiency.

White light emission can be obtained by mixing two complementary colors or three primary colors [2-

11]. Since highly efficient organic materials with green, yellow, and orange colors are available, it is most important to develop efficient blue emitting materials for developing efficient white OLEDs. Recently, Y.-H. Kim *et al.* [12] reported a very efficient, pure blue-emitting 9,10-bis[(2'',7''-di-*t*-butyl)-9',9''-spirobifluorenyl] anthracene (TBSA). TBSA has an absorption onset at 440 nm and a blue PL peak at 443 nm [12]. In this work, we report very efficient white OLEDs using TBSA as a blue light-emitting layer. The 4-dicyanomethylene-2-methyl-6-[2-(2,3,6,7-tetrahydro-1*H*,5*H*-benzo[*i,j*]quinolizin-8-yl)vinyl]-4*H*-pyran) (DCM2) was doped into a hole-transporting layer of 4,4'-bis[*N*-(1-naphthyl)-*N*-phenyl-amino]-biphenyl (α -NPD) to produce an orange color. The partial energy transfer from TBSA to DCM2 results in white light emission with two broad electroluminescence (EL) peaks around 470 (TBSA) and 580 nm (DCM2). By adjusting the DCM2 doping concentration and the thickness of the TBSA layer, we can fabricate white light-emitting devices with the Commission Internationale d'Éclairage (CIE) chromaticity coordinates of (0.32, 0.37). The external quantum efficiency is about 3.4 % and the luminous efficiency is about 3.9 lm/W at luminance of 100 cd/m².

2. Experimental

The devices were fabricated on precleaned ITO glass substrates with a sheet resistance of about 10 Ω/\square . To enhance the device performance, poly(3,4-ethylene dioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Baytron P4083) was deposited by spin coating from solution at 4000 rpm for 30 s, followed by drying at 100 °C for 1 hour in vacuum. Organic layers were deposited on top of the PEDOT:PSS layer by successive vacuum-deposition of α -NPD (500 Å), the red-emitting layer (50 Å), TBSA with various

thicknesses, Alq₃ (400 Å), LiF (5 Å), and Al electrode, without breaking vacuum. The red-emitting layer was deposited by simultaneously evaporating DCM2 and α -NPD with the DCM2 concentrations of 0.1, 0.2, and 0.3 %. For each DCM2 concentration in α -NPD, we fabricated devices with the TBSA thickness of 30, 60, 90 and 120 Å. We found that the TBSA thickness of 120 Å produces an appropriate blue EL intensity for the white light emission. The overlap area of the Al and ITO electrodes is about 2.5 mm². Fig. 1 shows the device structure and the molecular structure of TBSA and DCM2.

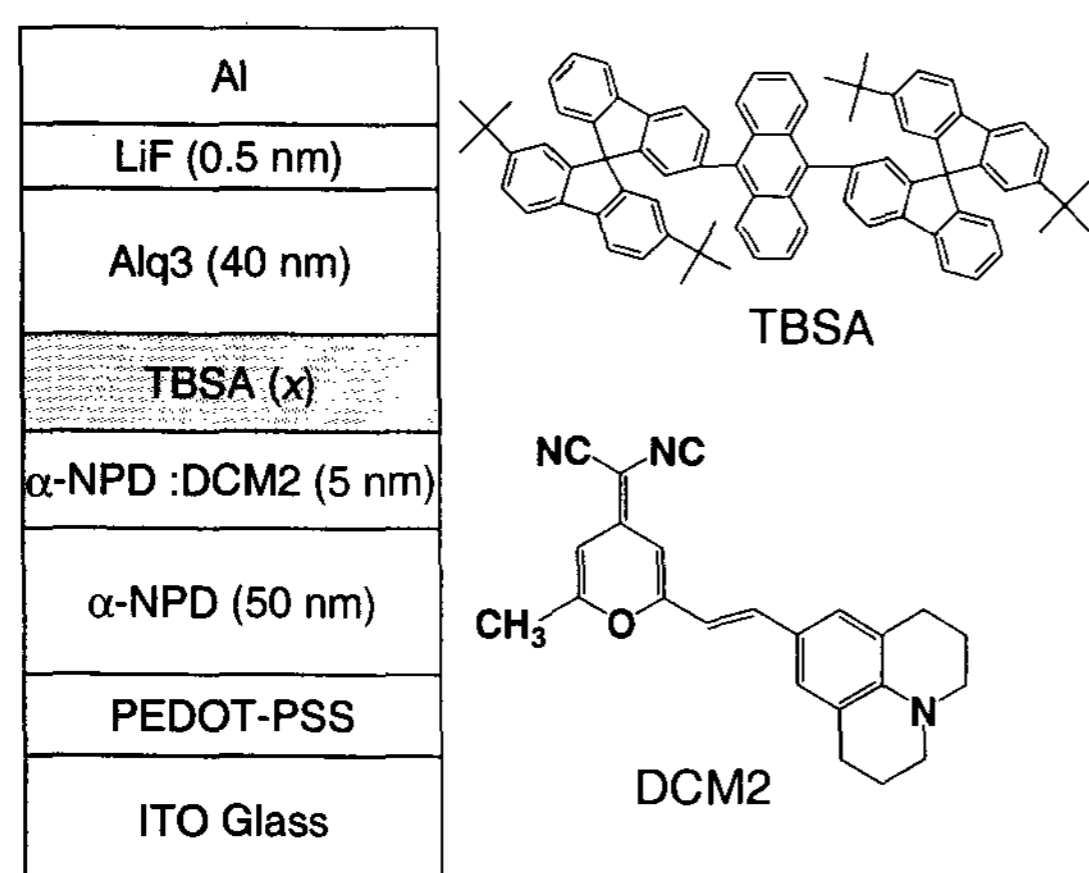


Figure 1. The device structure and the molecular structures of TBSA and DCM2.

The current-voltage-luminance (I-V-L) characteristics and the EL spectra were measured with a Keithley 236 source-measure unit and a Keithley 2000 multimeter equipped with a PMT through an ARC 275 monochromator. The external quantum efficiency (QE) of the EL, defined as the ratio of the emitted photons to the injected charges, was calculated from the EL intensity measured by the calibrated Si photodiode.

3. Results and discussion

Figure 2 compares the EL spectra measured under the current density of 20 mA/cm² for the devices with different DCM2 doping concentration in α -NPD. The thickness of the blue-emitting TBSA layer was 120 Å for all three devices. Figure 3 shows the CIE coordinates of the three devices; (0.25, 0.28), (0.32, 0.37) and (0.37, 0.39) for 0.1, 0.2 and 0.3 % DCM2 doped devices, respectively.

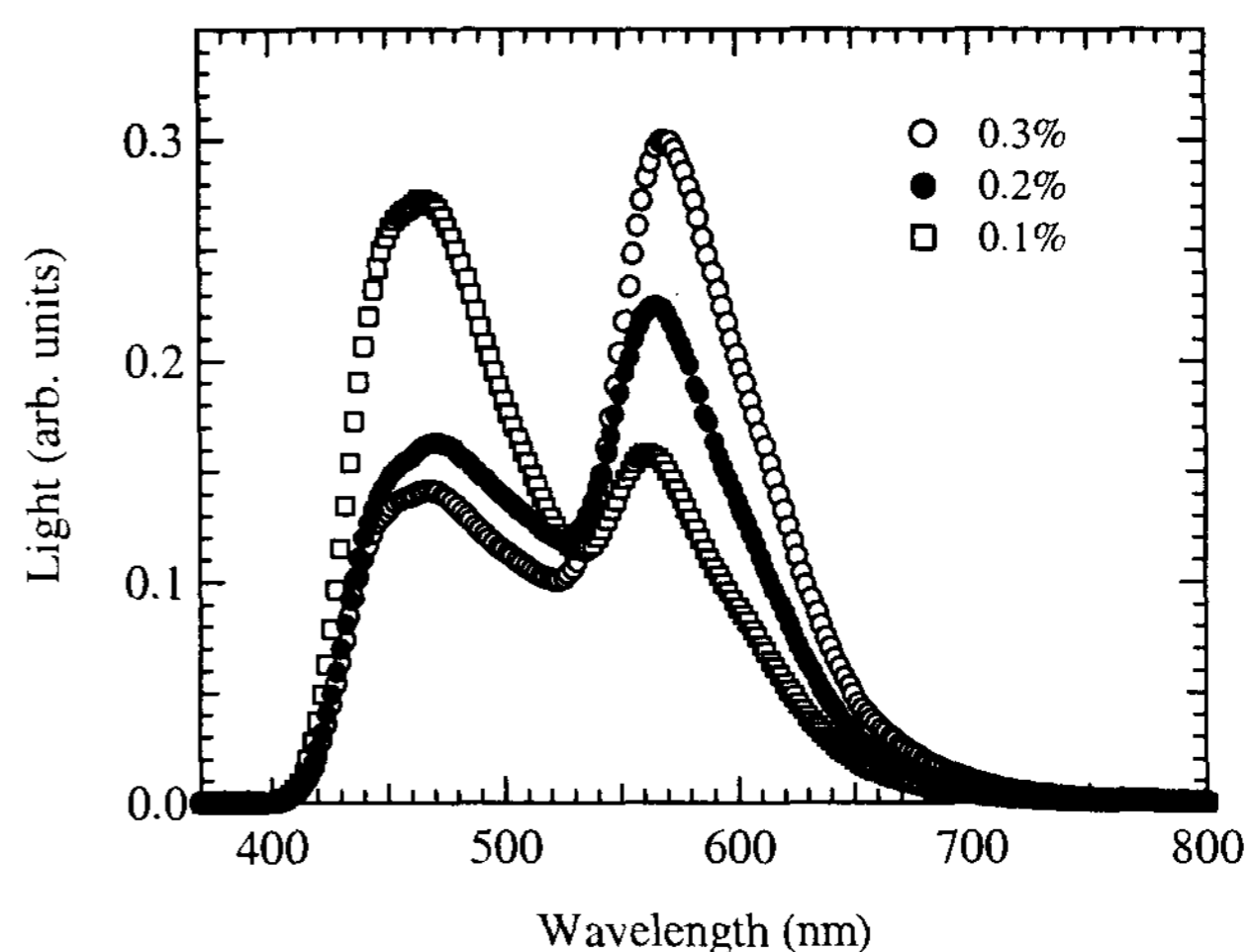


Figure 2. The EL spectra for devices of ITO/PEDOT:PSS/ α -NPD (50 nm)/ α -NPD:DCM2 (5 nm)/TBSA (12 nm)/ Alq₃ (40 nm)/LiF (0.5 nm)/Al at the current density of 20 mA/cm². Doping concentration of DCM2 in α NPD is 0.1 (open square), 0.2 (close circle) and 0.3% (open circle).

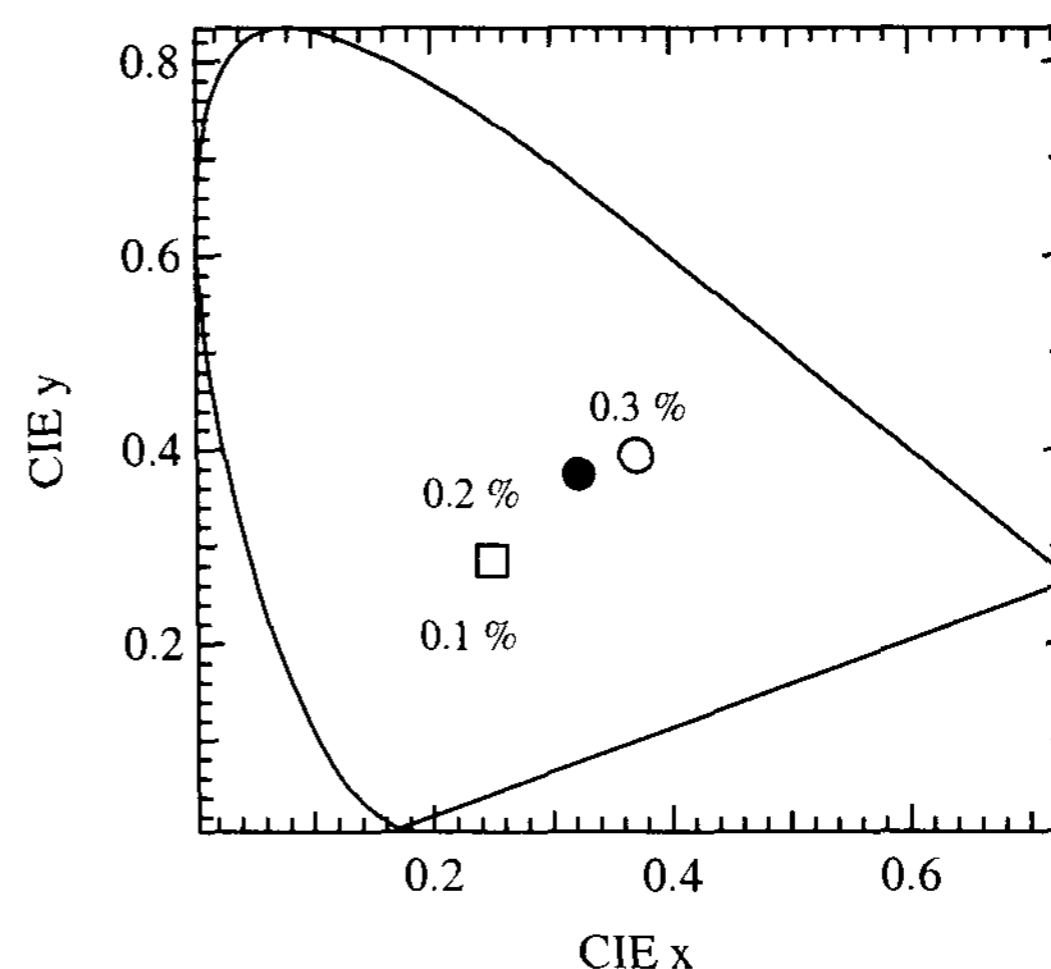


Figure 3. The CIE coordinates for the EL spectra in Fig. 2.

The EL spectra in Fig. 3 show a broad band covering the visible spectral range of 400–700 nm, with two peaks around 470 nm (TBSA) and 580 nm (DCM2), respectively. Since the hole-transport barrier is about 0.37 eV at the α -NPD/TBSA interface but the electron-transport barrier at the TBSA/Alq₃ interface is only 0.04 eV, the electron-hole recombination takes

place at the α -NPD/TBSA interface, resulting in the blue emission [12]. Fig. 3 shows that as the DCM2 concentration increases, the red emission of DCM2 increases and the blue peak of TBSA decreases. This result implies an energy transfer from TBSA to the nearby DCM2. Since the relative intensity of DCM2 with respect to TBSA increases with the doping concentration, it is possible to obtain a balanced white light by controlling the doping concentration of DCM2. The performances of three devices are summarized in Table 1. We found that the CIE coordinates are very close to the white point for the DCM2 concentration of 0.2 % and the TBSA thickness of 120 Å. This device shows a quite high external quantum efficiency of about 3.4 % and the luminous efficiency of about 3.9 lm/W at luminance of 100 cd/m².

Table 1. Performance of white organic light-emitting devices with different DCM2 concentrations in α -NPD.

DCM2 Conc. (%)	Max. Luminance (cd/m ²)	CIE _{xy}	Efficiency @ 100 cd/m ²	
			3.2 %	3.5 lm/W
0.1 %	38,800	(0.25, 0.28)	3.2 %	3.5 lm/W
0.2 %	45,400	(0.32, 0.37)	3.4 %	3.9 lm/W
0.3 %	55,800	(0.37, 0.39)	4 %	4.9 lm/W

Figure 4 shows the variation of the EL spectra with the current density for the device with the DCM2 concentration of 0.2 %, ITO/PEDOT:PSS/ α -NPD (50 nm)/ α -NPD:DCM2 (5 nm, 0.2 %)/TBSA (12 nm)/Alq₃ (40 nm)/LiF (0.5 nm)/Al. Although the red/blue ratio changes slightly with the current density, the overall EL spectra (and therefore the CIE coordinates) are quite stable for more than two orders of magnitude variation of the current densities. As the current density increases, the relative ratio of the red/blue emission decreases. This color shift may be understood as more hole injection into TBSA and the enhanced electron-hole recombination at the high electric field. Since the hole-transport barrier at the α -NPD/TBSA interface is quite high (\sim 0.37 eV), the hole injection into TBSA increases rapidly at the higher electric field, resulting in a larger blue emission from TBSA.

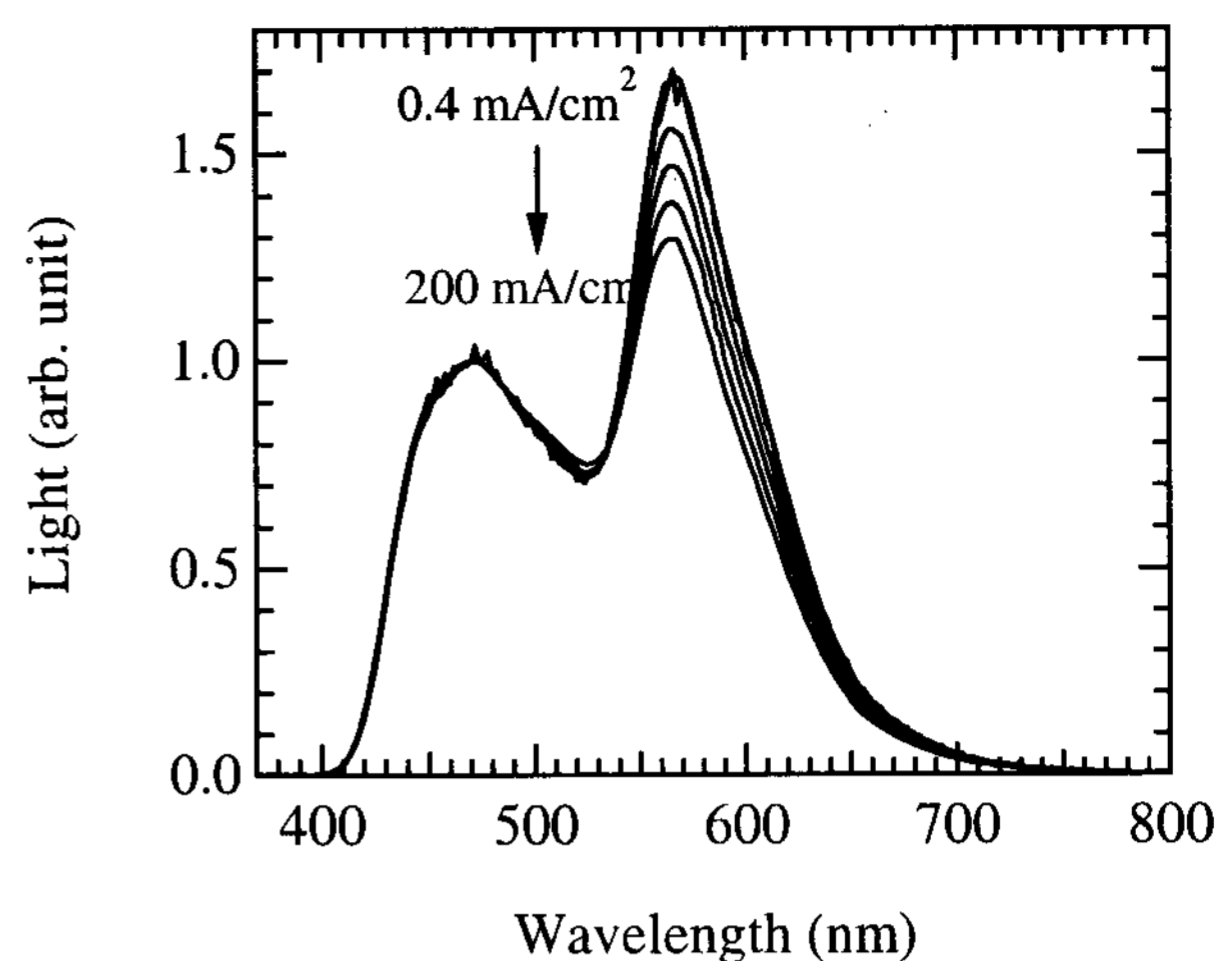


Figure 4. Variation of the EL spectra with increasing current density from 0.4 mA/cm² to 200 mA/cm² for a device of ITO/PEDOT:PSS/ α -NPD (50 nm)/ α -NPD:DCM2 (5 nm, 0.2 %)/TBSA (12 nm)/Alq₃ (40 nm)/LiF (0.5 nm)/Al.

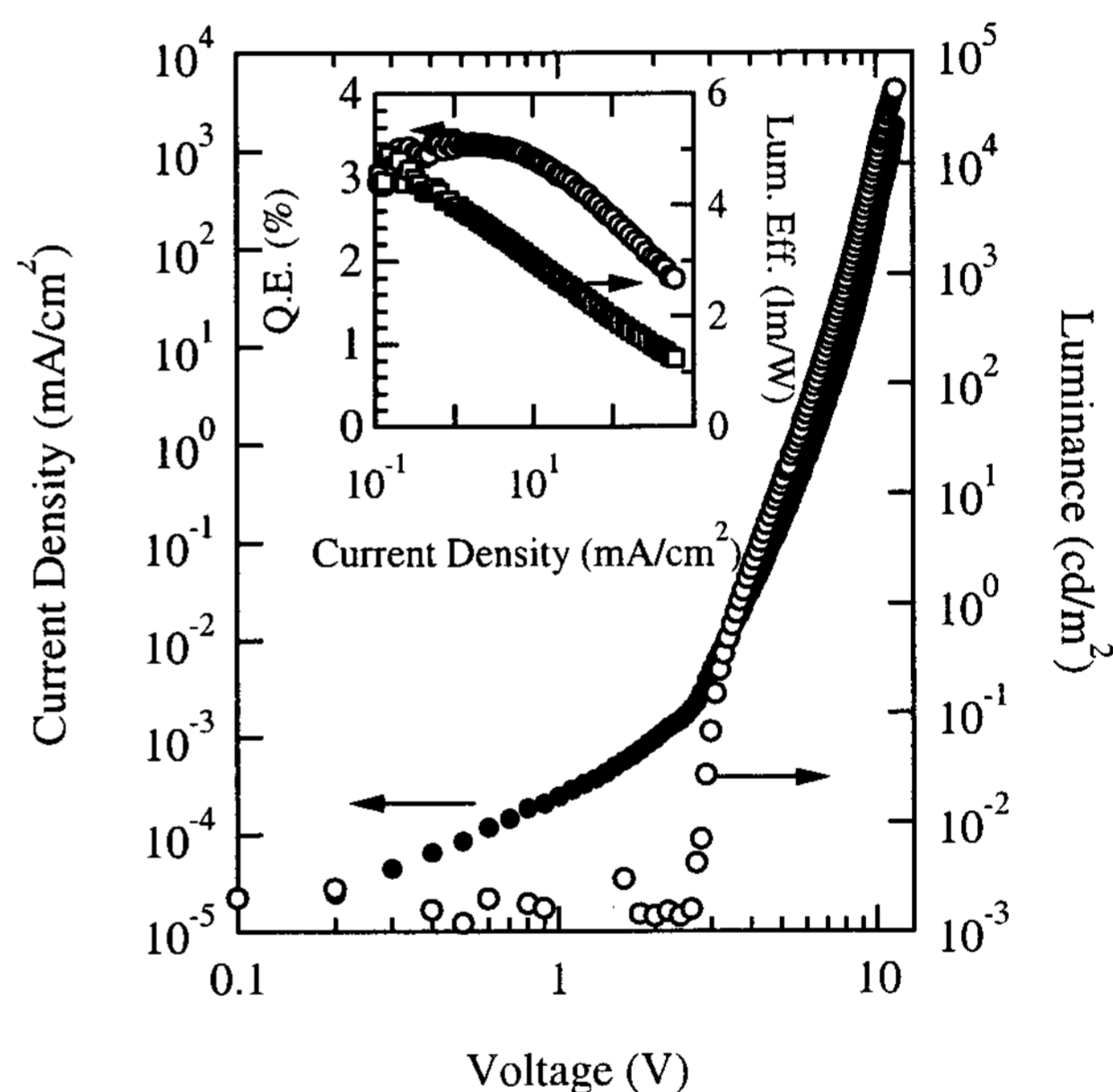


Figure 5. The current-voltage (solid circle) and luminance-voltage characteristics (open circle) of ITO/PEDOT:PSS/ α -NPD (50 nm)/ α -NPD:DCM2 (5 nm, 0.2 %)/TBSA (12 nm)/Alq₃ (40 nm)/LiF (0.5 nm)/Al. The inset shows an external quantum efficiency (QE) and the luminous efficiency as a function of the current density for the same device.

Figure 5 compares the current-voltage (I-V) and luminance-voltage (L-V) characteristics for ITO/PEDOT:PSS/ α -NPD (50 nm)/ α -NPD:DCM2 (5 nm, 0.2 %)/TBSA (12 nm)/Alq₃ (40 nm)/LiF (0.5 nm)/Al. The inset shows the external quantum efficiency (QE) and the luminous efficiency as the function of the current density for the same device. The onset voltage of the light emission is about 2.6 V. The device shows a maximum luminance of 45,400 cd/m² at 11.5 V and 1742 mA/cm². At a luminance of 100 cd/m² (current density of 1.31 mA/cm² and bias voltage of 6.5 V), the external quantum efficiency is 3.4 % and the luminous efficiency is 3.9 lm/W. The inset shows that the EL efficiency decreases with increasing current density. This phenomenon is attributed to the quenching of the singlet-excited state by a cationic species, which is common for devices with a dye doped emitting layer [13].

4. Conclusion

We reported very efficient white organic light-emitting devices consisting of two light-emitting layers of a blue-emitting TBSA layer and a red-emitting layer of DCM2 doped into α -NPD. The electron-hole recombination takes place at the α -NPD/TBSA interface, resulting in the blue emission. The partial energy transfer from TBSA to the nearby DCM2 produces white light emission with complementary colors of blue (TBSA) and orange (DCM2). By controlling the doping concentration of DCM2 and the TBSA thickness, we can obtain a balanced white light. We found that the CIE coordinates are very close to the white point for the DCM2 concentration of 0.2 % and the TBSA thickness of 120 Å. The device of ITO/PEDOT:PSS/ α -NPD (50 nm)/ α -NPD:DCM2 (5 nm, 0.2 %)/TBSA (12 nm)/Alq₃ (40 nm)/LiF (0.5 nm)/Al shows the CIE coordinates of (0.32, 0.37), the external quantum efficiency of about 3.4 % and the luminous efficiency of about 3.9 lm/W at luminance of 100 cd/m². The maximum luminance is about 45,400 cd/m² at 11.5 V.

5. Acknowledgements

This work was supported by the Brain Korea 21 Project in 2003 and funding from Advanced Backbone IT technology development project supported by Ministry of Information & Communication in republic of Korea.

6. References

- [1] C. W. Tang, S. A. VanSlyke, and C. H. Chen, *J. Appl. Phys.* **65**, 3610 (1989).
- [2] J. Kido, "Organic Electroluminescent Materials and Devices", Ed., S. Miyata and H. S. Nalwa (Gordon and Breach Science Publishers, Amsterdam, 1997), p. 335.
- [3] S. Tokito, J. Takata, Y. Taga, *J. Appl. Phys.* **77**, 1985 (1995).
- [4] J. Kido, M. Kimura, and K. Nagai, *Science* **267**, 1332 (1995).
- [5] R. H. Jordan, A. Dodabalapur, M. Strukelj, and T. M. Miller, *Appl. Phys. Lett.* **68**, 1192 (1996).
- [6] M. Granstrom, O. Inganas, *Appl. Phys. Lett.* **68**, 147 (1996).
- [7] R. S. Deshpande, V. Bulovic, and S. R. Forrest, *Appl. Phys. Lett.* **75**, 888 (1999).
- [8] C. W. Ko and Y. T. Tao, *Appl. Phys. Lett.* **79**, 4234 (2001).
- [9] C.-H. Kim, and J. Shinar, *Appl. Phys. Lett.* **80**, 2201 (2002).
- [10] J. T. Lim, N. H. Lee, Y. J. Ahn, G. W. Kang, C. H. Lee, *Current Appl. Phys.* **2**, 295 (2002).
- [11] G.-W. Kang, Y.-J. Ahn, J. T. Lim, and C. H. Lee, *Synth. Met.* **137**, 1029 (2003).
- [12] Y.-H. Kim, D.-C. Shin, S.-H. Kim, C.-H. Ko, H.-S. Yu, Y.-S. Chae, and S.-K. Kwon, *Adv. Mater.* **13**, 1690 (2001).
- [13] R. H. Young, C. W. Tang, and A. P. Marchetti, *Appl. Phys. Lett.* **80**, 874 (2002).