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White Organic Light-Emitting Diode Using the Exciplex Between DNTPD and ET-137

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Two organic layers of the N,N′-diphenyl-N,N′-bis-[4-(phenyl-m-tolyl-amino)-phenyl]-biphenyl-4,4′-diamine [DNTPD] and the ET-137 which is a pyran derivative provided from SFC Co. were sequentially deposited between the ITO anode and the LiF/Al cathode for generating blue light-emitting excitons in the electron transport material and yellow light-emitting exciplexes at the interface of two organic layers.

In this study, the DNTPD was used as a hole transport layer and the ET-137 as an electron transport and exciton formation layer. The structure of the fabricated devices is very simple compared with those of conventional white organic light-emitting devices (OLEDs) and the emission showed the nearly pure white on the Commission Internationale de I′ Eclarirage (CIE) chart.

The device with two organic layers of 500Å-thick DNTPD and 500Å-thick ET-137 showed electroluminescent characteristics of the double peak emission with a peak wavelength of 455 nm from the excitons of ET-137 and a peak wavelength of 561 nm from the exciplexes at the interface of DNTPD and ET-137, the maximum current efficiency of 1.8 cd/A, and the color coordinates of (0.32, 0.37) on the CIE chart.

Keywords DNTPD; EL peaks; ET-137; exciplex; exciton; white OLED

Introduction

White OLEDs have attracted much interest owing to their potential application in full color displays, backlights, and solid-state lightings. Conventional white OLEDs have generally three light components composed of red, green and blue [1–3] or two light components composed of blue and yellow [4,5]. The three color method has been considered as one of the best methods for generating white light, but it has low red emission efficiency so that the mixture of two colors is now considered as one of the viable technologies. In the multilayered white OLEDs, each layers according to the emission colors are composed of an energy-transferring host and a light-emitting dopant. In addition, lots of subsidiary layers for the injection, transport and confinement of carriers are required to achieve good electroluminescent characteristics so that the structure of white OLED becomes very complicated and a lot of organic materials are needed [6–8].

In this study, new white OLEDs with two organic layers of the DNTPD as a hole transport layer (HTL) and the ET-137 as an electron transport layer (ETL) as well as exciton

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formation layer were fabricated and their electroluminescent characteristics were evaluated according to the thickness combination of two organic layers. The proposed structure can be said to be the simplest of all the white OLEDs because the organic structure consists of only two kinds of materials without any dopants. The strategy to obtain the dual colors of blue and yellow for white emission is to use the blue emission from the excitons formed in the ET-137 region and the yellow emission from the exciplexes at the interface of DNTPD and ET-137.

Exciplex (excited state complex) is formed in the case in which a dimer between two organic materials is formed, that is, both components of the dimer are heterodimeric [9,10]. In the bilayer of DNTPD and ET-137, the exciplex is formed at the interface where there is a significant spatial overlap between the lowest unoccupied molecular orbitals (LUMOs) of the constituent species. When the exciplex returns to the ground state, its components dissociate and emit a light. The wavelength of an exciplex's emission is longer than those of the excited emissions of each species. Actually, electroluminescence (EL) spectra of a bilayer device usually depend on both the thickness of each organic layer and applied voltages [11]. Therefore, the nearly pure white emission can be obtained from an OLED with only two organic materials if the intensities of a blue emission from the excitons formed in one layer and a yellow emission from the exciplexes at the interface of two layers could be well mixed by the proper choice of kinds and thicknesses of the used materials.

In order to obtain good performance of white emission in a bilayer device using the exciplex, the thickness design as well as the proper choice of the used materials is very important because the large thickness of constituent species results in the reduction of luminance and current density due to the increase of series resistance and the small distance between electrodes and the organic interface causes poor electroluminescent characteristics due to the exciton quenching which leads to low quantum efficiency.

Experimental Procedure

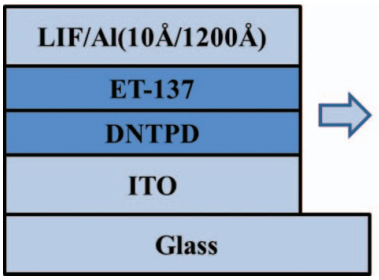
The substrates with an ITO (indium tin oxide) anode of 12 Ω/sq on glass were cleaned by ultrasonic cleaning process with acetone and isopropyl alcohol. The remaining solvent was removed by soft-baking for 10 minutes at 100°C. To improve the surface morphology of ITO transparent electrode film, the substrates were plasma-treated at 150W for two minutes under 8 mTorr pressure of O_2/Ar. The plasma treatment before the deposition of first organic layer reduces the energy barrier for hole injection from an anode and remove surface contaminants.

In the formation of DNTPD/ET-137, the deposition processes were divided into three kinds according to device classifications: the thicknesses of DNTPD/ET-137 were 300Å/500Å for the device A, 500Å/300Å for the device B, and 500Å/500Å for the device C as shown in Fig. 1. Finally, 10Å-thick LiF and 1200Å-thick Al were successively deposited as a cathode.

The electrical properties of the fabricated devices were measured using a Polaronix M6100 test system (McScience). The optical properties such as luminance, emission spectrum and CIE color coordinates were evaluated using a CS-1000 spectro-radiometer (Konica Minolta) in a dark room.

Results and Discussion

The energy diagram of the used materials is shown in Fig. 2. The DNTPD is well known as a HIL with a highest occupied molecular orbital (HOMO) level of 5.1 eV and a LUMO



Device	Configuration
A	DNTPD(300Å)/ ET-137(500Å)
B	DNTPD(500Å)/ ET-137(300Å)
C	DNTPD(500Å)/ ET-137(500Å)

Figure 1. Structure and classifications of the fabricated devices.

level of 2.1 eV in the energy diagram of Fig. 2 [12]. The holes from the anode of indium tin oxide (ITO) are easily injected into the DNTPD because its HOMO level is similar to Fermi level of ITO. The ET-137 as an electron transport and exciton formation layer has a HOMO level of 5.5 eV and a LUMO level of 2.6 eV. The zone of exciton formation is located in the ET-137 region near the interface because of the large LUMO and HOMO offsets at interface and high mobility of hole in organic materials. Therefore, the exciton emission is based on the singlets of ET-137 which emits a blue light.

Figure 3 shows EL spectra of the single layers of DNTPD and ET-137. The peak wavelengths in Fig. 3 were 427 nm for the DNTPD and 458 nm for the ET-137. The distance between the peak wavelengths of DNTPD and ET-137 is large enough to distinguish the emission of ET-137 from that of DNTPD. EL spectra of the fabricated devices were shown in Fig. 4, which were measured under an applied voltage of 7V and normalized to the blue peaks. The peak wavelengths of blue emission in Fig. 4 are nearly same with the peak wavelength of ET-137 in Fig. 3. This explains that the blue emission of a bilayer of DNTPD and ET-137 comes from the exciton emission of ET-137. On the other hand, the

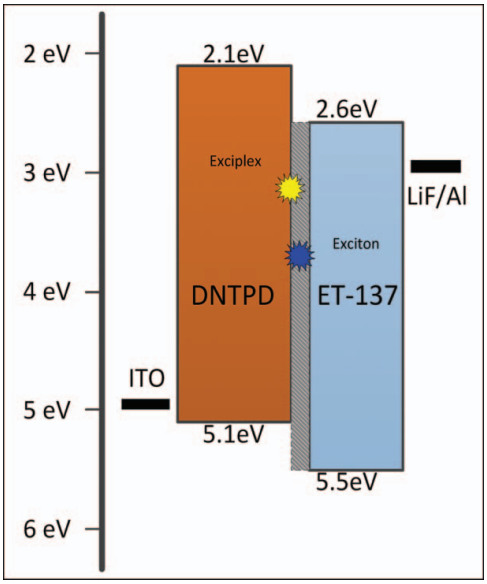


Figure 2. Energy diagram of the used materials.

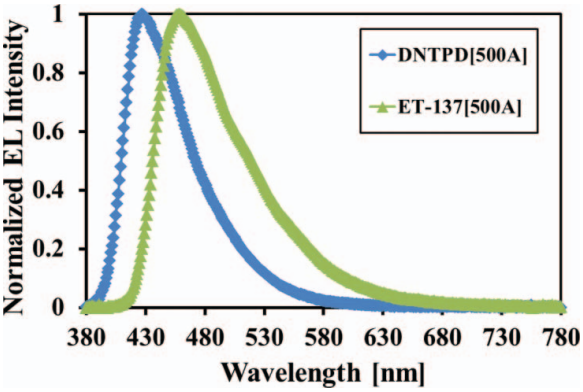


Figure 3. EL spectra of the single layers of DNTPD and ET-137.

peak wavelengths of yellow emission from the exciplexes slightly differs according to the fabricated devices because the exciplex emission depends on the thickness combination of two organic layers. The peak wavelengths of yellow emission are 556 nm for the devices (A, B) and 561 nm for the device C. As the yellow wavelengths of the fabricated devices are much longer than those of each layer of DNTPD and ET-137 in Fig. 3, we can know that the yellow emission in Fig. 4 was obtained from the exciplexes between DNTPD and ET-137. In the devices (A, B), the intensities of blue peaks are similar to those of yellow peaks, but the intensity of blue peak in the device C is higher than that of yellow peak. The relatively high intensity of blue to yellow peak in the device C compared to the devices (A, B) results from the reduction of exciton quenching due to the relatively long distance between electrodes and the exciton formation zone around the interface. The emission mixture of blue and yellow should be well contolled to obtain a pure white emission, but the intensities of yellow to blue emission in the devices (A, B) were rather high to obtain a good white color.

The current efficiency(η) can be calculated by the equation of $\eta = (\pi L)/(VJ)$ if the current density-voltage and luminance-voltage relationships are measured, where L (cd/m^2), V (V), and J (A/m^2) are luminance intensity, applied voltage, and current density,

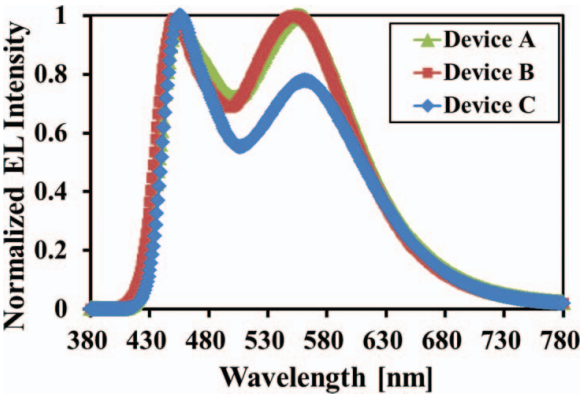


Figure 4. EL spectra of the fabricated devices under an applied voltage of 7 V.

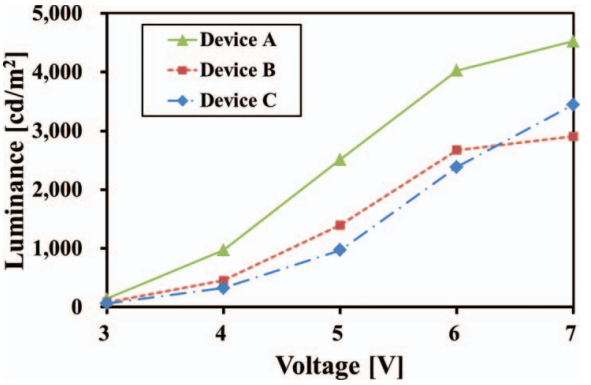


Figure 5. Luminance - voltage characteristics of the fabricated devices.

respectively. The luminance-voltage and current efficiency–luminance characteristics of the fabricated devices are shown in Figs. 5 and 6, respectively. The luminances under an applied voltage of 7 V were 4500 cd/m^2 for the device A, 2800 cd/m^2 for the device B and 3400 cd/m^2 for the device C in Fig. 5. The maximum current efficiencies were 2.0 cd/A for the device A, 1.1 cd/A for the device B, and 1.8 cd/A for the device C in Fig. 6. The luminance of device A is higher than that of device C because the device A has a higher current density due to a thinner layer of DNTPD. Comparing the electroluminescent characteristics of the devices (A, B), the luminance and current efficiency of device B are much lower than those of device A because the emission quenching can be more serious when the interface of two organic layers is nearer to the metal electrode than to the oxide electrode. The luminance of device B is even poorer than that of device C at a high voltage of 7V by emission quenching in spite of a thinner thickness of ET-137. Though the device A has the highest values of luminance and current efficiency among the experimental devices, it has a problem of color quality as a white emission. On the contrary, the emission color of device B shows nearly pure white with moderate values of luminance and current efficiency.

The CIE coordinates of the fabricated devices at 7V are shown in Fig. 7. The color coordinates were (0.32, 0.39) for the device A, (0.33, 0.38) for the device B, and (0.32, 0.37) for the device C. The emission color of device C was close to the pure white of (0.33,

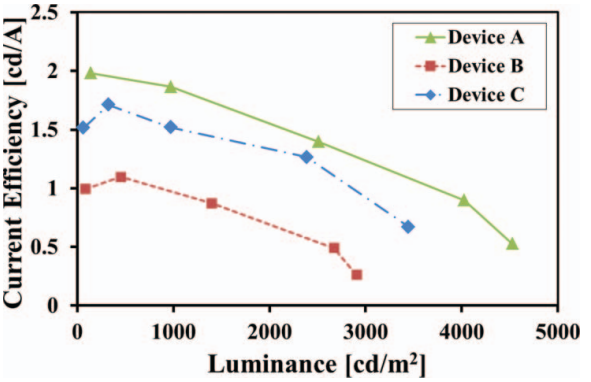


Figure 6. Current efficiency-luminance characteristics of the fabricated devices.

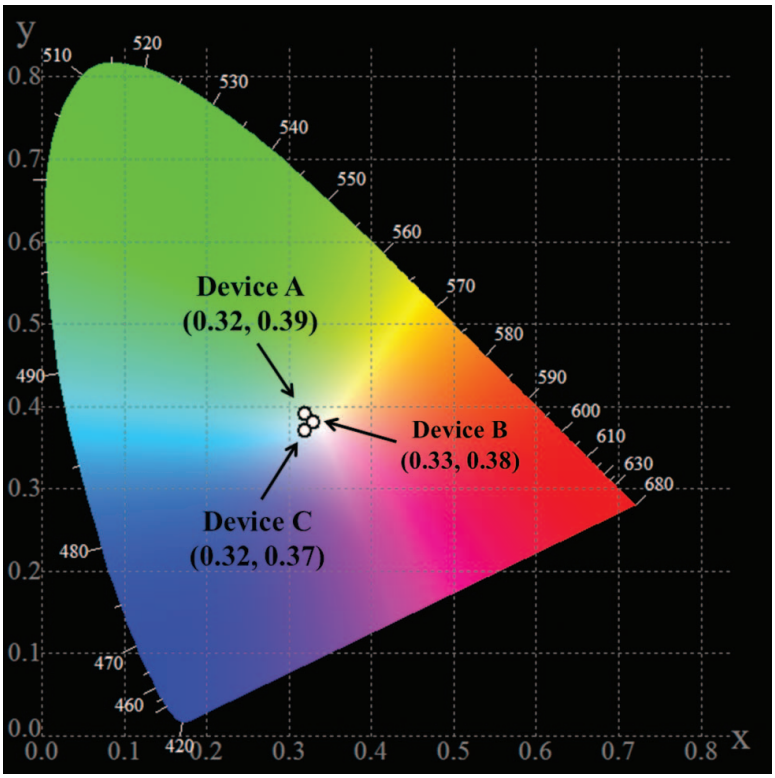


Figure 7. Color coordinates on the CIE chart.

0.33) on the CIE chart. In contrast, the emission colors of the devices (A, B) were rather yellowish-white due to the relatively strong yellow to blue emission.

Conclusions

New white OLEDs using two organic layers of DNTPD and ET-137 were fabricated and evaluated according to the thickness combination of DNTPD and ET-137. The nearly pure white emission could be obtained in the device C with two organic layers of 500Å-thick DNTPD and 500Å-thick ET-137. In the structure of two organic layers, the DNTPD was used as a hole transport layer and the ET-137 as an electron transport and exciton formation layer.

The proposed structure is very simple compared with those of conventional white organic light-emitting devices (OLEDs) due to the use of only two materials without any dopants. The strategy to obtain the dual colors of blue and yellow for white emission is to use both blue emission from the excitons formed in the ET-137 region and yellow emission from the exciplexes at the interface of DNTPD and ET-137.

In the device C, the maximum current efficiency was 1.7 cd/A and the luminance and color coordinates at 7V were 3400 cd/m^2 and (0.32, 0.37), respectively. And also, EL spectra showed the double peak wavelengths of 455 nm and 561 nm.

The good white emission in the device C could be obtained from the well-balanced mixture of blue and yellow intensities.

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