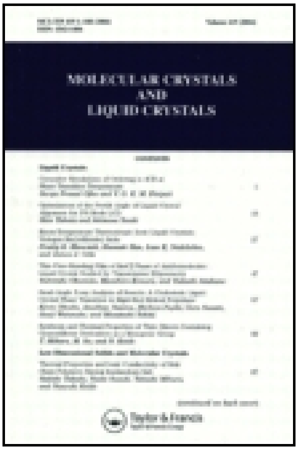

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Electroluminescence Mechanism for the Efficiency Enhancement and the Color Stabilization of Yellow Organic Light-Emitting Devices with a Mixed Layer

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The luminous efficiency of the organic light-emitting devices (OLEDs) with a mixed layer in a hole transport layer (HTL) and an emitting layer (EML) was highest among the fabricated OLEDs, and the luminous efficiency of the OLEDs with a mixed layer at $10\,mA/cm^2$ *was* $6.5\,cd/A$. *The Commission Internationale de l'Eclairage (CIE) chromaticity coordinates of the OLEDs fabricated utilizing a mixed layer became stabilized, and the CIE chromaticity coordinates of the OLEDs at 15 V was (0.457, 0.510), indicative of a deep yellow color. The efficiency enhancement and the color stabilization of the OLEDs with a mixed layer were attributed to the exciplex emission process.*

Keywords: electroluminescence mechanism; luminous efficiency; organic light-emitting devices

PACS numbers: 78. 60. Fi and 78. 66. Qn

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I. INTRODUCTION

Organic light-emitting devices (OLEDs) have been particularly interesting because of their potential applications in flat-panel displays, which have unique advantages of high contrast, fast response, low power consumption, and wide-view angle [1–6]. The typical emitting process of the OLEDs requires to inject holes of the anode and electrons of the cathode into the organic layers, to form excitons resulting from the electron-hole recombination and to emit the light by the radiative decay of the excitons [7–9]. When the OLEDs with several layers have been fabricated, an excited state complex (exciplex) related to the luminescence is formed by the intermolecular interaction between molecules at organic layer heterointerfaces, blends, or mixed layers [10–13]. The exciplex formation significantly generates due to a spatial overlap between the lowest unoccupied molecular orbital (LUMO) of the constituent species [14]. Since the efficiency and the color stabilization of yellow OLEDs are significantly affected by the existence of a mixed layer, systematic studies on the electroluminescence (EL) mechanism in the OLEDs with a mixed layer are very important for improving their efficiencies and achieving high performance.

This paper reports for the EL properties of OLEDs with various structures fabricated by using organic molecular-beam deposition (OMBD). Photoluminescence (PL) and EL measurements were carried out to investigate the optical properties and the color stabilities of the OLEDs with a mixed layer. EL mechanism for the efficiency enhancement and the color stabilization of OLEDs with a mixed layer in the emitting layer (EML) and the hole transport layer (HTL) are described on the basis of the EL results.

II. EXPERIMENTAL DETAILS

The sheet resistivity and the thickness of the indium-tin-oxide (ITO) thin film used in this study were 30 Ω/square and 1000 Å. The ITO substrates were cleaned using sonications of acetone, methanol, and distilled water at 60°C for 15 min. After the cleaned ITO substrates are dried by using N_2 gas with a purity of 99.9999%. The surfaces of the ITO substrates were treated with oxygen plasma for 2 min at an O_2 pressure of approximately 2×10^{-2} Torr. The five kinds of samples used in this study were deposited on ITO thin films coated on glass substrates by using OMBD and consisted of the following structures from the top: an aluminum (Al) cathode electrode, a lithium quinolate (Liq) electron injection layer (EIL), a tris-(8-hydroxyquinoline) aluminum (Alq_3) EML/electron transport layer (ETL), a perylene,

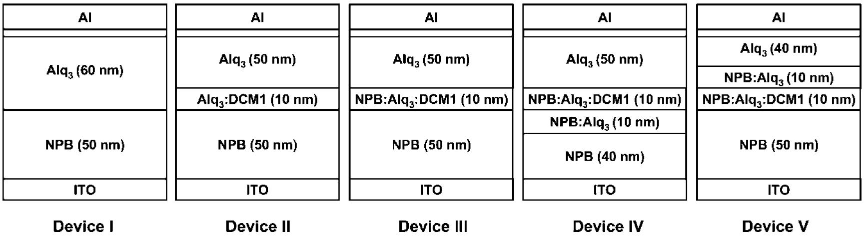

FIGURE 1 Schematic diagrams of the fabricated OLEDs with various structures used in this study. The mixed ratio consists of NPB and Alq_3, and the ratio of the deposition rate of the NPB layer to that of Alq_3 layer is 1:1. The doping concentration of the DCM1 molecule in the Alq_3 emitting layer is 1%.

4-dicyano-methylene-2-methyl-6-4-dimethylami-nostyryl-4H-piran (DCM1) yellowish dopant (1%) [15], a *N,N*′-diphenyl-*N,N*′-bis(1-napthyl)-(1,1′-biphenyl)-4,4′-diamine (NPB) HTL, an ITO anode electrode, and glass substrates. While the thickness of the Alq_3 layer and the NPB layer for the OLEDs were different, the distance from the ITO anode to the DCM1 dopant interface for hole injection or the distance from the Al cathode to the DCM1 dopant interface for electron injection were maintained at constant value of 50 nm. The organic materials were evaporated onto an ITO coated glass substrate at a chamber pressure of about 3×10^{-6} Torr. The growth rates of the organic layers and the metal layers were approximately 0.01 Å/s and 1 Å/s, respectively. After organic and metal depositions, the OLED devices were encapsulated in the glove box with O_2 and H_2O concentrations below 1 ppm. The size of emitting region in the pixel is $5 \times 5\,mm^2$. The brightness was measured by using a brightness meter, chroma meter CS-100A (Minolta). All measurements were conducted at room temperature in ambient environment. The EL spectra were obtained using a LS50B (Perkin Elmer) EL spectrometer. Schematic diagrams of the OLEDs with various structures are shown in Figure 1.

III. RESULTS AND DISCUSSION

Figure 2 show (a) a ultra-violet (UV) absorption spectrum and (b) a PL spectrum. The excitation source for the PL measurements is a UV lamp. The dominant peak around 570 nm for the PL spectrum appears, and the full width at half maximum (FWHM) of the corresponding peak is approximately 64 nm. The PL peak around 570 nm is related to the DCM1 solution. The absorption peak for the UV

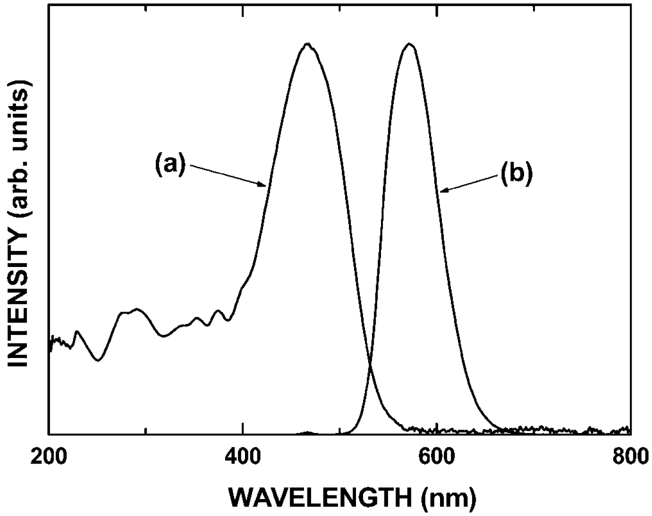

FIGURE 2 (a) Ultra violet absorption spectrum and (b) photoluminescence spectrum for the DCM1 solution in methylene chloride.

absorption spectrum appears at 470 nm, which is the maximum wavelength of the excitation source.

The EL spectra at 14 V for OLEDs with various structures are shown in Figure 3. The EL spectrum of device II shows two emission peaks. One dominant peak at 570 nm is attributed to the DCM1 dopant as shown in Figure 2, and a shoulder at 510 nm is related to the Alq_3 layer. Another dominant EL peak at 550 nm for devices III, IV, and V might be related to the exciplex emission in a mixed EML between NPB and Alq_3 materials, as yet unidentified line. The intensity of the shoulder corresponding to the Alq_3 layer is affected by the magnitude of the carrier mobility due to the NPB:Alq_3 mixed layer. Even though the carrier injection efficiency into the EML for the device III is better than devices IV and V, a small intensity corresponding to the Alq_3 layer shown in Figure 3 originates from the imbalance of the hole and the electron in the EML, resulting in the maximum luminance of 13200 cd/m^2 at 15 V. The NPB:Alq_3 mixed layer acting as a HTL in the device IV optimizes the balance between the hole and the electron in the NPB:Alq_3:DCM1 EML, resulting in the enhancement of the efficiency of the OLEDs, which is a maximum luminance of 15500 cd/m^2 at 15 V, and in disappearance of the Alq_3 emission peak [16]. However, the electron mobility of the NPB:Alq_3 mixed layer acting as an ETL in device V significantly decreases due

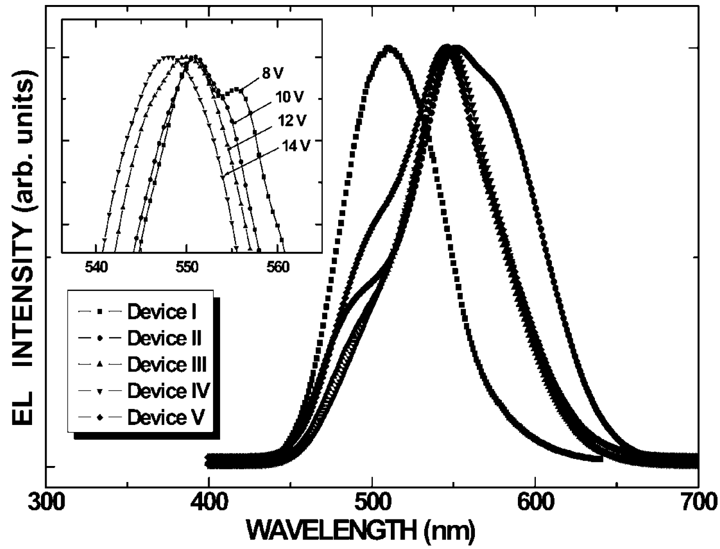

FIGURE 3 Electroluminescence spectra at 14 V for OLEDs with various structures. The inset indicates the blue shifts of the electroluminescence spectra for device IV at 8, 10, 12, and 14 V.

to an increase in the hopping distance of the electron in the mixed layer. When the emission region of the OLEDs taking into account the Alq_3 emission peak component transfers from the NPB:Alq_3:DCM1 EML to the NPB:Alq_3 mixed layer, the maximum luminance at 15 V is 1880 cd/m^2. The energy level difference between the highest occupied molecular orbitals (HOMOs) of the NPB and the Alq_3 layers (0.4 eV) is smaller than that of the corresponding LUMOs (0.65 eV), the holes in the NPB layer inject into the Alq_3 layer and recombine with electrons, resulting in the formation of exciples in the mixed layer. Therefore, the hole mobility in the HTL is moderately decreased due to an increase in the hopping distance of the hole, resulting in the efficiency enhancement of the exciplex emission within the NPB:Alq_3:DCM1 EML. Among the five kinds of OLEDs, device IV has the highest efficiency, which is 6.4 cd/A at 10 mA/cm^2. Furthermore, the FWHM of the EL spectrum for device IV is as narrow as 70 nm. The luminance efficiencies for devices I, II, III, IV, and V are summarized in Table 1.

The Commission Internationale de l'Eclairage (CIE) chromaticity coordinates at 15 V of the devices show that the CIE coordinates of devices III, IV, and V with a mixed layer shift to a pure yellow color with increasing applied voltage. The CIE coordinates for devices I,

TABLE 1 Luminance Efficiencies at 10 mA/cm^2 and CIE Coordinates at 15 V for Devices I, II, III, IV, and V

Device	Luminance efficiencies (cd/A)	CIE coordinates (x, y)
I	2.8	(0.350, 0.550)
II	5.4	(0.477, 0.492)
III	4.9	(0.462, 0.513)
IV	6.4	(0.458, 0.516)
V	5.4	(0.457, 0.510)

II, III, IV, and V are summarized in Table 1. The CIE chromaticity coordinates of the OLEDs with a mixed layer at 15 V was (0.457, 0.510), indicative of a deep yellow color.

Figure 4 shows EL mechanism of the yellow color emission related to the exciplex emission process in the device IV. The exciplex process originates from the charge transfer interaction between the excited/ground state of the donor level and the ground/excited state of the acceptor level. The exciplex process under electrical excitation is related to the confinement of the carriers injected from the opposite electrodes between donor and acceptor molecules [17]. The EL mechanism of the device IV is investigated to clarify the efficiency

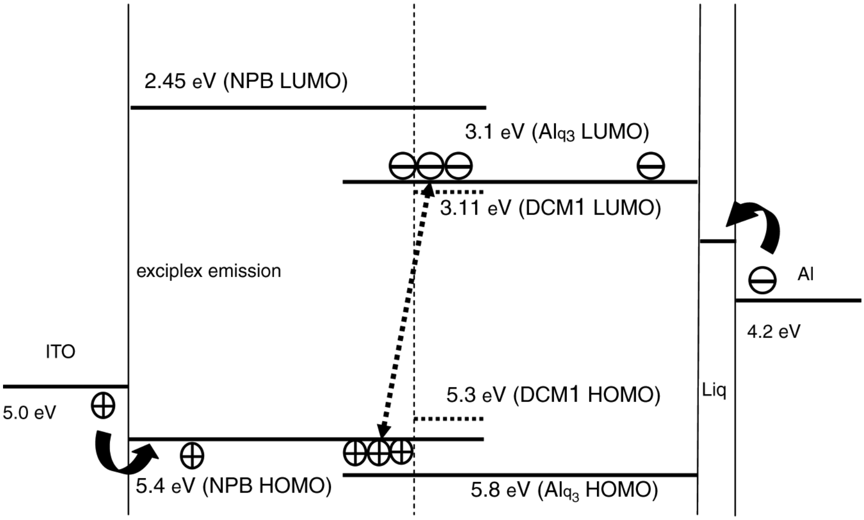

FIGURE 4 Electroluminescence mechanism of the yellow color emission related to the exciplex emission between holes in the NPB molecules and electrons in the Alq$_3$ molecules in the device IV.

enhancement and the color stabilization. The indirect recombinations of accumulated holes within the HOMO level of the NPB layer and the depleted electrons within the LUMO level of the Alq_3 layer in the NPB:Alq_3:DCM1 EML are more dominant, resulting in the formation of the NPB:Alq_3 exciplex. The dominant exciplex emission process is give by (1)

$$h\omega_{ex} = E_{LUMO}(Alq_3) - E_{HOMO}(NPB), \tag{1}$$

where $E_{LUMO}(Alq_3)$ is the LUMO of Alq_3 (−3.1 eV), E_{HOMO}(NPB) is the HOMO of NPB (−5.4 eV), and ω_{ex} is the frequency of the exciplex emission. An exciplex emission wavelength of the mixed organic materials (545 nm), determined by using the Eq. (1), is in reasonable agreement with the experimental value (550 nm) shown in Figure 3. Therefore, the EL peak at 550 nm for device IV shown in Figure 3 is attributed to the exciplex emission process resulting in the efficiency enhancement.

When an operating voltage increases, a dominant exciplex emission peak for device IV shows blue shift behavior, as shown in the insert of Figure 3. Even though the EL peak of a device with a guest impurity doped host layer typically shows red shift behavior with increasing dopant concentration due to the solid state solvation effect [18,19], the peak of device IV depicts blue shift behavior with increasing voltage due to a decrease in the domain of the host polarity resulting from the addition of the NPB molecules into the DCM1 doped Alq_3 layer. Therefore, an appropriate regulation of the host polarity may provide the possibility for controlling precisely the color tuning [21,21].

IV. SUMMARY AND CONCLUSIONS

The EL properties and mechanisms of yellow OLED structure were investigated. The OLEDs with a mixed layer acting as a HTL and an EML showed the highest efficiency among the samples, and the CIE coordinates of the OLEDs at 15 V was (0.457, 0.510), indicative of a deep yellow color. The efficiency enhancement and the color stabilization of the OLEDs with a mixed layer were attributed to the exciplex emission process. These present observations can help improve understanding of the EL mechanisms for the efficiency enhancement the color stabilization of yellow OLEDs with a mixed layer.

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