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Enhancement of the Color Stabilization in Yellow Organic Light-Emitting with N, N'-bis-(1-naphthyl)-N, N'-diphenyl-1,1'-biphenyl-4,4'-diamine/5,6,11,12-tetraphenylnaphthacene Multiple Heterostructures Acting as an Emitting Layer

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Enhancement of the Color Stabilization in Yellow Organic Light-Emitting with N, N'-bis-(1-naphthyl)-N, N'-diphenyl-1,1'-biphenyl-4,4'-diamine/5,6,11, 12-tetraphenylnaphthacene Multiple Heterostructures Acting as an Emitting Layer

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The optical properties of organic light-emitting devices (OLEDs) with multiple heterostructures consisting of N, N'-bis-(1-naphthyl)-N, N'-diphenyl-1,1'-biphenyl-4,4'-diamine(NPB)/5,6,11,12-tetraphenylnaphthacene (rubrene) emitting layers were investigated. The optical properties of the OLEDs were significantly affected by the number of heterostructures. The electroluminescence intensity corresponding to the rubrene peak for the OLEDs increased with increasing driving voltage and with increasing number of heterostructures. The Commission Internationale de l'Eclairage (CIE) chromaticity coordinates became stabilized with increasing the number of heterostructures, and the coordinates maintained almost constant, regardless of the driving voltage. The CIE chromaticity coordinate of the OLEDs with 5-periods of the heterostructures was (0.37, 0.54), indicative of a yellow color.

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I. INTRODUCTION

Potential applications of organic light-emitting devices (OLEDs) in promising next-generation full-color flat-panel displays have driven extensive efforts to enhance brightnesses and efficiencies [1–6]. OLED displays are particularly attractive because of their promising applications, which offer various advantages of low driving voltage, low power consumption, high contrast, wide viewing angle, low cost, and fast response [7,8]. Even though many studies on enhancing OLED efficiencies have been performed, OLEDs still have inherent problems due to limited self-luminescence [9–12], low efficiency [13,14], short lifetime [15,16], and poor color stability [17], resulting from the degradation of the OLEDs. Potential applications of OLEDs have driven extensive efforts to overcome these inherent problems utilizing fabricating novel OLED structures. Among the novel OLED structures, OLEDs using multiple heterostructures have been introduced to obtain narrower spectral emissions, higher emission efficiencies, and tunable emission spectra [18–21]. Recently, high-efficiency OLEDs with a tunable light emission were achieved by using aromatic diamine/5,6,11,12-tetraphenylnaphthacene (rubrene) multiple heterostructures [20]. Even though some works on efficiency enhancement of the OLEDs by using organic multiple heterostructures have demonstrated improved balance of holes and electrons in the emitting layers [18–21], systematic studies concerning the color stability OLEDs utilizing multiple heterostructures have not reported yet because of the complicated device-fabrication process.

This article reports the optical properties of OLEDs utilizing multiple heterostructures deposited by using organic molecular-beam deposition (OMBD). Electroluminescence (EL) measurements were carried out to investigate the optical properties of the Aluminum (Al)/lithium quinolate (Liq)/tris(8-hydroxyquinoline) aluminum (Alq₃)/multiple heterostructures/N, N'-bis-(1-naphthyl)-N, N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB)/indium-tin-oxide (ITO)/glass structures with and without multiple heterostructures. The multiple heterostructures were consisted of both NPB and rubrene. The dependence of the optical properties on the heterostructure number in the Al/Liq/Alq₃/multiple heterostructures/NPB/ITO/glass structures was investigated, and the Commission Internationale de l'Eclairage (CIE) chromaticity coordinates corresponding to the emission color were determined.

II. EXPERIMENTAL DETAILS

The sheet resistivity of the ITO thin films coated on glass substrates used in this study was $30\ \Omega/\text{sq.}$. The ITO substrates were cleaned using acetone and methanol at 60°C for 5 min, and rinsed in de-ionized water thoroughly. After the chemically cleaned ITO substrates had been dried by using N_2 gas with a purity of 99.9999%, the substrates were treated with oxygen plasma for 10 min. The three kinds of samples used in this study were deposited on ITO thin films coated on glass substrates by using OMBD with effusion cells and shutters and consisted of the following structures from the top: an Al (100 nm) cathode electrode, a Liq (2 nm) electron injection layer, an Alq_3 (60 nm) electron transport layer, no, a 3-period, or a 5-period NPB/rubrene multiple heterostructure emitting layer, a NPB (10 nm) hole transport layer, an ITO anode electrode, and glass substrates. The total thickness of the NPB/rubrene multiple heterostructures was approximately 40 nm. The thicknesses of the NPB and the rubrene layers in the OLEDs with 3 periods of multiple heterostructures were 10.3 and 3 nm, respectively, and the corresponding values in the OLEDs with 5 periods of multiple heterostructures were 5 and 3 nm, respectively. The NPB/rubrene multiple heterostructure emitting layer is used to enhance the OLED efficiency due to the existence of the rubrene molecules, which act as a hole trap in the OLEDs [4,22]. The Liq layer is used as an electron injection layer, leading to a lower turn-on voltage and higher power efficiency [23]. The depositions of the OLED layers were done at a substrate temperature of 27°C and system pressure of 5×10^{-8} Torr. The growth rates of the organic layers and the metal layers were approximately 0.1 and $0.5\ \text{\AA}/\text{s}$, respectively. The OLEDs without NPB/rubrene multiple heterostructures were grown for comparison with the OLEDs with NPB/rubrene multiple heterostructures.

III. RESULTS AND DISCUSSION

The schematic diagrams of the Al/Liq/ Alq_3 /multiple heterostructures/NPB/ITO/glass structures (a) without multiple heterostructures and with (b) 3 and (c) 5 periods of multiple heterostructures are shown in Figure 1. The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) levels of the rubrene are -5.4 and $-3.2\ \text{eV}$, respectively [24], and the HOMO and the LUMO levels of the NPB layer, as obtained by using cyclic voltammetry, are -5.5 and $-2.5\ \text{eV}$, respectively [24]. Electron or hole mobilities in organic materials are related with the drift velocity of carriers to the applied electric

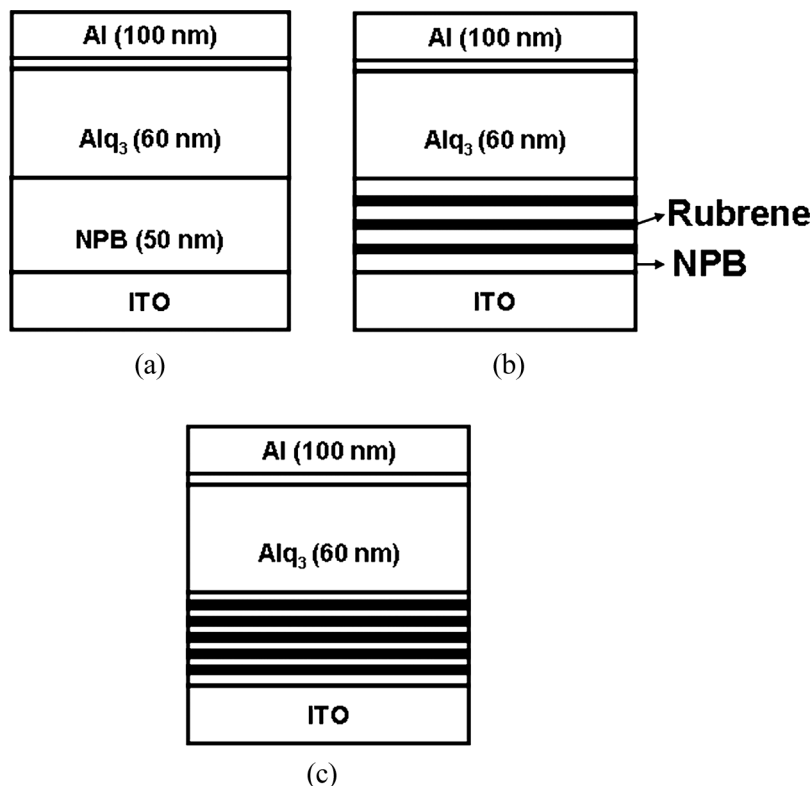


FIGURE 1 Schematic diagrams of the Al/Liq/Alq₃/multiple heterostructures/NPB/ITO/glass structures (a) without multiple heterostructures and with (b) 3 and (c) 5 periods of multiple heterostructures.

field. Since holes are accumulated and trapped in the HOMO levels of the NPB/rubrene quantum wells of the OLEDs due to the existence of the barrier at the NPB/rubrene heterointerface, the mobility of the holes in the multiple heterostructures is decreased, which enhances the efficiency of the OLEDs by achieving a better balance between the numbers of electrons and holes [4]. Because of this improved balance, the recombination probability in the NPB/rubrene multiple heterostructures and at the NPB/Alq₃ heterointerface increases.

Figure 2 shows the EL spectra at 12 V for the Al/Liq/Alq₃/multiple heterostructures/NPB/ITO/glass structures (a) without multiple heterostructures and with (b) 3 and (c) 5 periods of multiple heterostructures. The dashed lines indicate the decomposition of the EL spectra. Figure 2(a) shows a dominant peak at 510 nm for the EL spectrum of

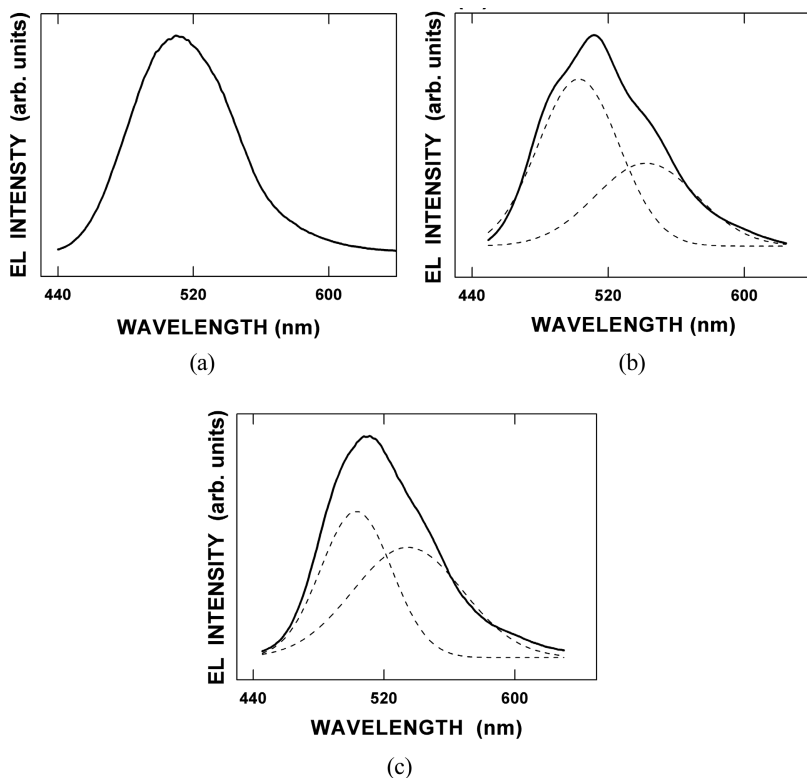


FIGURE 2 Electroluminescence spectra at 12 V for the Al/Liq/Alq₃/multiple heterostructures/NPB/ITO/glass structures (a) without multiple heterostructures and with (b) 3 and (c) 5 periods of multiple heterostructures. The dashed lines indicate the decomposition of the electroluminescence spectra.

the OLEDs without multiple heterostructures, which is related to the Alq₃ layer. The appearance of the emission peak related to the Alq₃ layer is attributed to the significant creation of the exciton recombination resulting from the existence of the electron and the hole in the Alq₃ layer. The EL characteristics of the combined rubrene and Alq₃ emission components are shown in Figures 2(b) and 2(c). Figures 2(b) and 2(c) show that the EL intensity corresponding to the rubrene in the OLEDs increases with increasing number of the multiple heterostructures and that the peak intensity corresponding to the rubrene in the OLEDs with 5 periods of multiple heterostructures is much larger than that of the OLEDs with 3 periods of multiple heterostructures. The full width at half maximum (FWHM) at 12 V of

the EL spectrum for the OLEDs with 3 and 5 periods of multiple heterostructures are 77 and 72 nm, respectively. The FWHMs of the EL spectra for the OLEDs with 5 periods of multiple heterostructures studied in this work were narrower than those for the OLEDs using aromatic diamine/rubrene in the emitting layer [20]. For OLEDs with 3 periods of multiple heterostructures, the intensity of the decomposed rubrene peak is much smaller than that of the decomposed Alq₃ peak while for OLEDs with 5 periods of multiple heterostructures, the intensity of the decomposed rubrene peak is much closer to that of the decomposed Alq₃ peak. The blue shift of the emission peak related to the Alq₃ layer at 480 nm shown in Figures 2(b) and 2(c) in comparison with the peak in Figure 2(a) might be attributed to the appearance of the excimer emission or the exciplex emission in the rubrene/NPB heterointerface. Thus, the

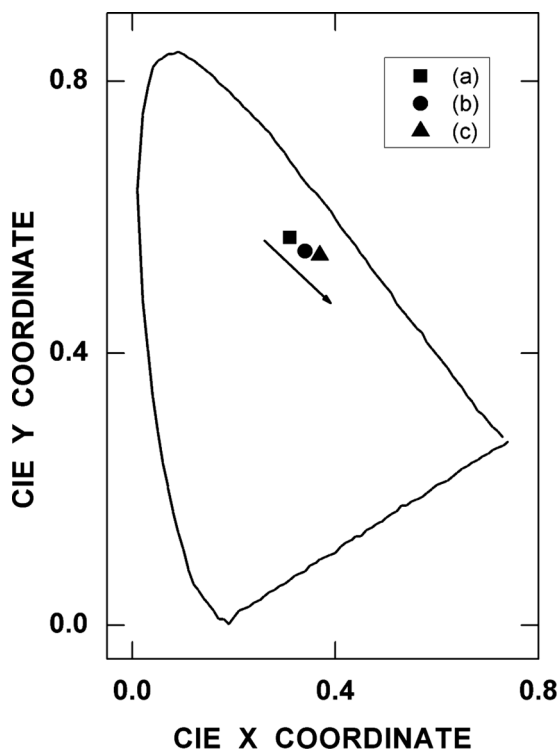


FIGURE 3 Commission Internationale de l'Eclairage coordinates at 12V for the Al/Liq/Alq₃/multiple heterostructures/NPB/ITO/glass structures (a) without multiple heterostructures and with (b) 3 and (c) 5 periods of multiple heterostructures.

FWHM of the EL peak corresponding to the rubrene decreases with increasing number of multiple heterostructures. The EL peak corresponding to the rubrene shifts to low wavelength side with increasing the number of the heterostructures due to the hole distribution and confinement in the multiple heterostructures [20]. However, the EL peak related to the Alq_3 remains the same position, regardless of the number of the heterostructures. While the EL peak related to the rubrene in the OLEDs with 3 periods of multiple heterostructure appears, that in the OLEDs with co-evaporation of the NPB and rubrene does not appear, indicative of a decrease of the carrier confinement in the rubrene layer.

The CIE coordinates at 12 V for the Al/Liq/ Alq_3 /multiple heterostructures/NPB/ITO/glass structures (a) without multiple heterostructures and with (b) 3 and (c) 5 periods of multiple heterostructures are (0.31, 0.57), (0.34, 0.55), and (0.37, 0.54), respectively, as shown in Figure 3. The coordinates for the OLED with 5 periods of multiple heterostructures correspond to a stabilized yellow color, which is emitted from the rubrene layer. Figure 4 shows the CIE coordinates as functions of the driving voltages for the Al/Liq/ Alq_3 /multiple heterostructures/NPB/ITO/glass structures (a) without multiple heterostructures and

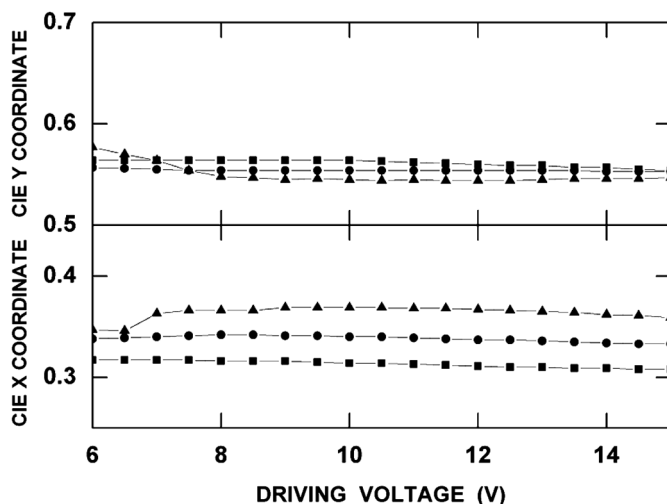


FIGURE 4 Commission Internationale de l'Eclairage coordinates as functions of the driving voltage for the Al/Liq/ Alq_3 /multiple heterostructures/NPB/ITO/glass structures with various numbers of multiple heterostructures. Filled rectangles, filled circles, and filled triangles represent the OLEDs without and with 3 and 5 periods of multiple heterostructures, respectively.

with (b) 3 and (c) 5 periods of multiple heterostructures. The CIE coordinates become stabilized with increasing the number of heterostructures, and the coordinates maintain almost constant, regardless of the driving voltage. The intensity ratio of the Alq₃ emission and the rubrene emission decreases with increasing driving current density. This behavior is attributed to the shift of the dominant emission zone with increasing driving current density.

IV. SUMMARY AND CONCLUSIONS

The optical properties of the OLEDs with NPB/rubrene multiple heterostructures were investigated. While the turn-on voltage of the OLEDs increased with increasing number of multiple heterostructures due to an increase in the number of trapped holes, the efficiency of the OLEDs was enhanced with increasing number of multiple heterostructures due to improved balance of holes and electrons in the emitting layer. The optical properties of the OLEDs were significantly affected by the existence and the number of multiple heterostructures. The CIE chromaticity coordinates became stabilized with increasing the number of heterostructures, and the coordinates maintained almost constant, regardless of the driving voltage. The CIE chromaticity coordinates of the OLEDs with 5-periods of the multiple heterostructures were (0.37, 0.54), indicative of a yellow color. These results indicate that the color stabilized OLEDs fabricated with multiple heterostructures hold promise for potential applications as yellow sources in full-color displays.

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