Efficiency enhancement mechanism in organic light-emitting devices with multiple heterostructures acting as a hole transport layer

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Abstract

The electrical and the optical properties of organic light-emitting devices (OLEDs) with or without multiple heterostructures acting as a hole transport layer were investigated. The efficiency enhancement mechanism in the OLEDs with multiple heterostructures is described on the basis of the electrical and the optical results.

1. Introduction

Organic light-emitting devices (OLEDs) have been very attractive due to their being promising candidates for next-generation flat-panel displays, which offer various advantages of low driving voltage, low power consumption, high contrast, wide viewing angle, low cost, and fast response [1]. Because the mobility of holes in a hole transporting layer (HTL) in OLEDs is a few orders of magnitude higher than that of the electrons in an electron transporting layer (ETL) [2], a decrease in the number of holes in the HTL or an increase in the electron injection into the ETL is very important for increasing electron-hole recombination in an emitting layer (EML) resulting in an improved OLED efficiency. Therefore, studies of charge injection from electrodes to organic materials and charge transport in organic layers are necessary for enhancing the efficiencies and the lifetimes of OLEDs. Even though some works concerning enhancements of the efficiencies and the lifetimes of OLEDs utilizing various structures have been performed [3], few studies concerning the efficiency enhancement mechanism in OLEDs with multiple heterostructures acting as a HTL have been reported.

The paper reports the electrical and the optical properties of OLEDs with multiple heterostructures acting as a HTL deposited by using organic molecular-beam deposition (OMBD). The multiple heterostructures consisted of both N, N′-bis-(1-naphthyl)-N, N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) and 4,4',4"-tris(N-3-methylphenyl-N-phenyl-amino)-triphenylamine (m-MTDATA). Current density-voltage (J-V), luminance-voltage, efficiency-current density, and electroluminescence (EL) measurements were carried out to investigate the electrical and the optical properties of the OLEDs with and without NPB / m-MTDATA multiple heterostructures.

2. Experimental

The indium-tin-oxide (ITO) thin films with a sheet resistivity of 20 Ω/□ coated on glass were used as the substrates in this study. The ITO substrates were cleaned using sonications of acetone, methanol, and distilled water at 60 for 5 min, and rinsed in de-ionized water thoroughly. The chemically cleaned substrates were kept for 48 h in isopropyl alcohol. After the chemically cleaned substrates had been dried by using N_2 gas with a purity of 99.9999%, the surface of the substrates were treated with an oxygen plasma for 10 min at an O_2 pressure of approximately

2×10^{-2} Torr. The four kinds of samples used in this study were deposited on ITO thin films coated on glass substrates by using OMBD with tungsten effusion cells and shutters and consisted of the following structures from the top: an Al cathode electrode, a Liq EIL, DCM1 selectively doped Alq_3, either no, a 3-period or a 5-period m-MTDATA/NPB multiple heterostructures, an ITO thin films coated on glass substrates.

The NPB/m-MTDATA multiple heterostructure HTL is used to enhance the OLED efficiency due to the existence of the m-MTDATA molecules. The depositions of the OLED layers were done at a substrate temperature of 27°C and a system pressure of 5×10^{-8} Torr. The growth rate of the organic layers and the metal layers were approximately 1 and 10 /s, respectively, which were controlled by using a quartz crystal thickness monitor. After organic and metal depositions, the OLED devices were encapsulated in a glove box with O_2 and H_2O concentrations below 1 ppm. A desiccant material consisting of barium-oxide powder was used to absorb the residue moisture and oxygen in the encapsulated device. The size of the emitting area in the pixel was 5 × 5 mm^2. All measurements were conducted at room temperature in ambient environment. The J-V characteristics of the OLEDs were measured on a programmable electrometer with built-in current and voltage measurement units (model 236, Keithely). The brightness was measured by using a brightness meter, chroma meter CS-100A (Minolta), and the EL spectra were obtained using a LS50B (Perkin Elmer) EL spectrometer. The four kinds of the OLEDs fabricated in this study are described in Table. I.

3. Results and discussion

Table I. OLEDs with various structures used in this study. The doping concentration of the DCM1 molecule in the emitting layer is 0.7%.

Device / Layer	I	II	III	IV
Cathode (nm)	Al (100)			
EIL (nm)	Liq (2)			
EML (nm)	Alq_3 (60)	Alq_3 (40) / 0.7% DCM1 doped Alq_3 (5) / Alq_3 (15)		
HTL (nm)	NPB (50)		{NPB (15) /[m-MTDATA(3.3) /NPB (5)]$_3$/ NPB (10)}	{NPB (15) /[m-MTDATA (2) / NPB (2.5)]$_5$ / NPB (12.5)}
Anode	ITO			

Figure 1 shows schematic energy band diagrams of the OLEDs without and with a NPB/m-MTDATA multiple heterostructures acting as a HTL. The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) levels of the m-MTDATA are -5.11 and -1.91 eV, respectively [4], and the HOMO and the LUMO levels of the NPB layer, as obtained by using cyclic voltammetry, are -5.46 and -2.45 eV, respectively [5]. Since holes are accumulated and trapped in HOMO levels of the NPB/m-MTDATA multiple heterostructures of the OLEDs due to the existence of the barrier at the NPB/m-MTDATA heterointerface, the mobility of the holes in the multiple heterostructure is decreased, which enhances the luminance efficiency of the OLEDs by achieving a better balance between the numbers of electrons and holes. The decrease of the hole mobility in the multiple heterostructure will be clarified in the EL spectra. The recombination probability at the NPB/Alq_3 heterointerface increases due to this improved electron-hole balance.

Figure 2(a) shows the J-V characteristics of the OLEDs without and with the multiple heterostructures. The J-V characteristic of device I shows similar behavior to that of device II, indicative that the carrier transport of the OLEDs is not significantly affected by the DCM1 layer with a doping concentration of 0.7 wt%. Because the concentration and the thickness of the DCM1 molecules are very low (0.7%) and thin (5nm), respectively, they cannot affect significantly the carrier transport in OLEDs. Because a decrease in the current density of devices III and IV is attributed to the decrease of the hole mobilities, resulting in more holes being accumulated multiple heterostructures, the current densities of devices I and II without multiple heterostructures are higher than those of the devices III and IV with multiple heterostructures. The luminance efficiency-current density characteristics show that a maximum efficiency of 6.779 cd/A at 16.02 mA/cm^2 for device III was obtained, as seen in Fig. 2(b). The improvement of the efficiency is related to a better electron-hole balance in the EML.

The EL spectrum of device I shows a dominant peak at 506.5 nm corresponding to an Alq_3 layer. The dominant EL peak position for the OLEDs with a DCM1-doped Alq_3 layer is 591 nm [6]. If the recombination zone moves to the interface between the NPB and the Alq_3 layers due to the decrease of

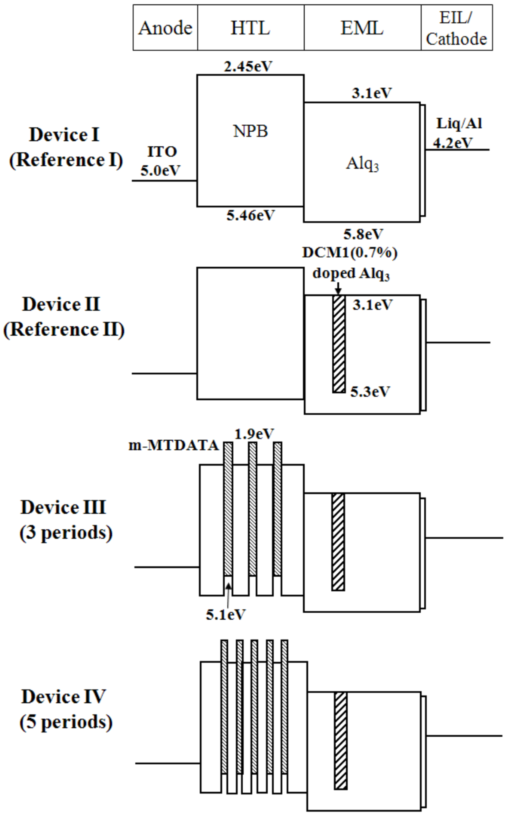

Fig. 1. Schematic diagrams of the fabricated OLEDs without or with multiple heterostructures used in this study. The LUMO and the HOMO represent the lowest unoccupied molecular orbital and the highest occupied molecular orbital, respectively. The doping concentration of a DCM1 is 0.7 wt%.

hole mobility, the intensity of the EL peak corresponding to the DCM1 is decreased. The EL intensities of devices III and IV at 591 nm are smaller than that of device II, indicating that the recombination probability is reduced in the DCM1-doped Alq_3 layer. Because the hole mobilities of devices III and IV are decreased due to the insertion of NPB/m-MTDATA multiple heterostructures, the recombination zone in EML moves to the interface between the HTL and the EML. The decrease of the hole mobility in the multiple heterostructures is clarified from the position of the recombination zone.

4. Summary

The OLEDs with multiple heterostructures acting as a HTL showed the highest luminances and efficiencies due to an increase in the recombination probability, resulting from the decrease of the hole mobility. The EL intensities of devices III and IV at 591 nm were smaller than that of device II, indicating that the recombination probability was reduced in the DCM1-doped Alq_3 layer. The movement of the recombination zone in the OLEDs with multiple heterostructures due to the decrease in the hole mobility, which was clarified by the EL spectra.

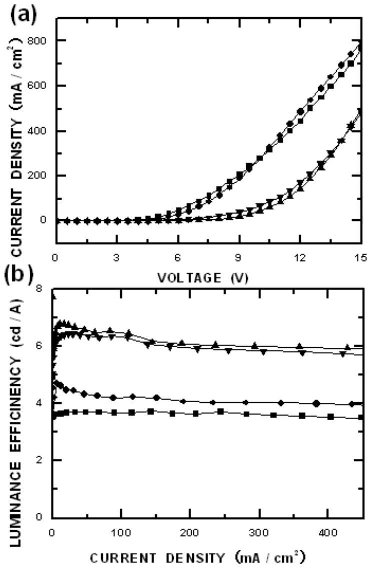

Fig. 2. (a) Current densities as functions of applied voltages and (b) efficiencies as functions of current densities for the OLEDs without and with multiple heterostructures. Filled rectangles, filled circles, filled up-triangles, and filled down-triangles represent the OLEDs of devices I, II, III, and IV, respectively.

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5. References

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