Enhancement of efficiencies for tandem green phosphorescent organic light-emitting devices with a *p*-type charge generation layer

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ABSTRACT

Tandem green phosphorescent organic light-emitting devices with a 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile or a molybdenum trioxide charge generation layer were fabricated to enhance their efficiency. Current density–voltage curves showed that the operating voltage of the tandem green phosphorescent organic light-emitting device with a 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile layer was improved by 3% over that of the corresponding organic light-emitting device with a molybdenum trioxide layer. The efficiency and the brightness of the tandem green phosphorescent organic light-emitting device were 13.9 cd/A and 26,540 cd/m^2, respectively. The current efficiency of the tandem green phosphorescent organic light-emitting device with a 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile layer was lower by 1.1 times compared to that of the corresponding organic light-emitting device with molybdenum trioxide layer due to the decreased charge generation and transport in the 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile layer resulting from triplet–triplet exciton annihilation.

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1. Introduction

Organic light-emitting devices (OLEDs) have emerged as excellent candidates for potential applications in mobile displays, flat panel displays, and lighting sources and they have excellent advantages of low-power consumption, fast response time, thinness, high contrast ratio, and low-cost production. However, the efficiency of OLEDs is still relatively low in comparison with those of other commercialized display devices, such as cathode ray tubes and liquid crystal displays [1–3]. Tandem OLEDs have attracted a great deal of interest for enhancing the current efficiency, luminance, lifetime, and tunable emission spectrum. A charge generation layer (CGL), which supplies electrons and holes to the emitting layer (EML) in each OLED, is essential for enhancing the efficiency of tandem OLEDs. When a *p*-type CGL is used as a hole-injecting conductive layer, holes are injected into the upper OLED, and the CGLs improve the carrier transport efficiency due to a decrease in the charge barrier. The high efficiencies for tandem OLEDs are attributed to the CGL with a highly-transparent metal layer [4,5]. Organic hole-transporting layers doped with a Lewis

acid [6] or with *p*-type metal oxides, such as V$_2$O$_5$, WO$_3$, or MoO$_3$, have been used as CGLs [7–15]. Studies concerning tandem OLEDs with inorganic CGLs have been extensively performed to achieve high brightness and current efficiency. However, the inorganic materials are unstable due to chemical reactions, short lifetimes, and evaporation at the high growth temperatures [16,17]. Even though some studies on the fabrication and the device characteristics of green phosphorescent OLEDs have been performed, very few studies concerning the enhancement of efficiencies for tandem green phosphorescent OLEDs with a *p*-type CGL have been conducted.

This paper reports data on the enhancement of the efficiencies for tandem green phosphorescent OLEDs with a *p*-type 1,4,5,8,9,11-hexaazatriphenylene-hexacarbonitrile (HAT-CN) or a MoO$_3$ CGL. Current density–voltage, luminance–voltage, and luminance efficiency–current density curves, as well as electroluminescence (EL) spectra, were measured to investigate the device characteristics and the luminance mechanisms of the OLEDs with a HAT-CN or a MoO$_3$ CGL.

2. Experimental

The sheet resistivity of the indium-tin-oxide (ITO) thin films deposited on glass substrates used in this study was 15 Ω/square. The ITO-coated glass substrates were cleaned in a solution of acetone and methanol for 10 min by using an ultrasonic cleaner

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and were rinsed in de-ionized water. After the ITO substrates had been chemically cleaned, they were dried by using N_2 gas with a purity of 99.9999%. The cleaned ITO substrates were treated with UV O_3 for 10 min at room temperature and a system pressure of 1 atm. The prepared ITO substrates were introduced into the evaporation chamber, and the organic layers and the metal layer were deposited on the ITO substrates at a substrate temperature of 28 °C and a system pressure under 1.2×10^{-4} Pa. The deposition rates of the organic layer and the metal electrode, which were controlled by using a quartz crystal deposition rate/thickness monitor (Sigma, SQM-160), were 1–2 Å/s and 1.5–2 Å/s, respectively. The thicknesses of the hole injection layer (HIL) and of the CGLs with a HAT-CN layer or a MoO_3 layer were optimized by using a hole-only device and a charge generation current device. The hole-only device and the charge generation current device consisted of ITO (150 nm)/HAT-CN (x nm)/N,N'-bis(naphthalene-1-yl)-N,N'-bis(phenyl)-benzidine (NPB) (40 nm)/4,4',4''-tris(N-3-methylphenyl-N-phenylamino) triphenylamine (m-MTDATA) (20 nm)/NPB (40 nm)/Al (100 nm) and ITO (150 nm)/8-tris-hydroxyquinoline (Alq$_3$) (300 nm)/HAT-CN or MoO_3 (x nm)/NPB (40 nm)/Al (100 nm). The green phosphorescent tandem OLEDs contained the structure NPB/1,3-bis(cabazol-9-yl) benzene (mCP): fac-tris(2-phenylpyridine) iridium (Ir(ppy)$_3$) (8 wt%)/Alq$_3$, which is denoted as a single OLED unit, and had the following three structures: ITO/HAT-CN/single OLED unit/single OLED unit/8-hydroxy-quinolinato lithium (Liq)/Al, ITO/HAT-CN/single OLED unit/MoO_3 CGL/single OLED unit/Liq/Al, and ITO/HAT-CN/singleOLED unit/HAT-CN CGL/single OLED unit/Liq/Al, which are denoted as devices I, II, and III, respectively. Schematic diagrams of the tandem structures and the corresponding energy band diagrams for devices I, II, and III are shown in Fig. 1(a), (b), and (c), respectively.
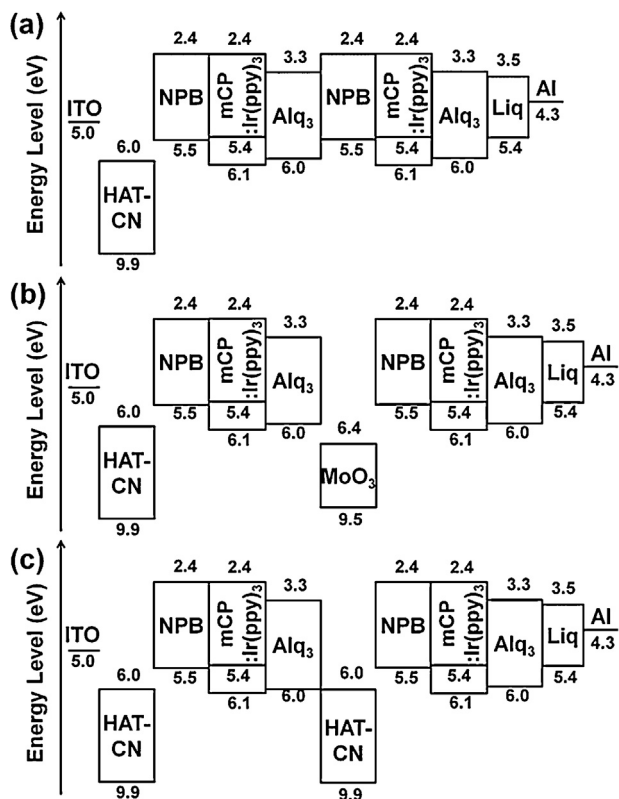
The current–voltage–luminance characteristics and the EL spectra of the green OLEDs were measured in a dark box by using a current–voltage measurement system (M6100, McScience) and a spectroradiometer (CS-1000A, Minolta). Currents, luminances, and EL spectra were simultaneously measured by applying bias voltages to the tandem green phosphorescent OLEDs in a dark box under ambient conditions. The luminance efficiencies were calculated by using the measured luminances, current densities, and voltages.

3. Results and discussion

Hole-only devices with an m-MTDATA electron blocking layer (EBL) were fabricated to investigate hole transport and injection, as shown in Fig. 2. The EBL interrupts the electron flow through the organic layer; thus, the carriers flowing through the organic layer are dominantly attributed to hole injection and transport. The HAT-CN layer acts as a hole injection layer (HIL), resulting in a decrease in the hole injection barrier due to the ITO and the NPB layers with very high electron affinities. The thickness of the HAT-CN HIL for the hole-only device was optimized at 5 nm. The voltages of the hole-only devices with a 5 nm HAT-CN layer that reached current density of 10 mA/cm^2 was reduced by 27% compared to those of the hole-only devices without a HAT-CN layer.

No current was observed in the charge generation current device without a CGL under forward bias, as shown in Fig. 3, indicative of the existence of an Alq$_3$ hole-blocking layer and an NPB electron-blocking layer, whereas charge generation current devices with a HAT-CN layer or a MoO_3 layer between the Alq$_3$ and the NPB layers exhibited a current response [18]. The optimal thicknesses of the HAT-CN and the MoO_3 CGLs for the charge generation current devices were significantly related to the inner carrier current generated due to the existence of the Alq$_3$ and the NPB layers that had been deposited as an inverted structure to block carrier injection from both electrodes. The optimized thicknesses of the MoO_3 and the HAT-CN CGLs for the charge generation current devices were 3 and 5 nm, respectively. The operating voltages of the charge generation current devices with a MoO_3 layer and with a HAT-CN layer at 10 mA/cm^2 were 11.2 and 10.6 V, respectively. Thus, the operating voltage of the charge generation current device with a HAT-CN layer was about 5.4% smaller than that of the device with a MoO_3 layer.

The operating voltages of devices I, II, and III at 10 mA/cm^2 were 21, 17, and 16.5 V, respectively, as shown in Fig. 4(a). The operating
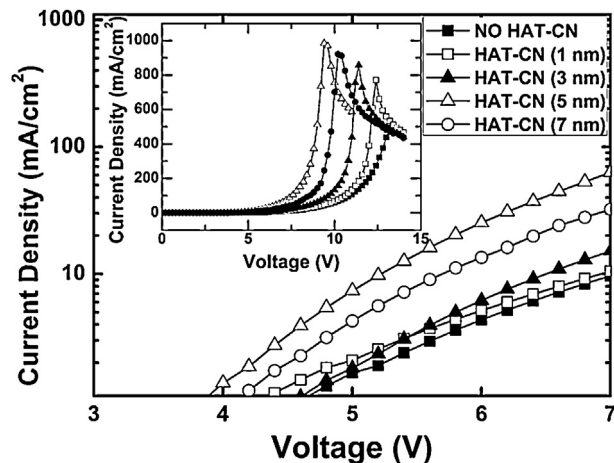
Fig. 1. Schematic energy band diagrams of the OLEDs (a) without a charge generation layer and with (b) a MoO_3 charge generation layer or (c) a HAT-CN charge generation layer.

Fig. 2. Current densities as functions of the applied voltage for hole-only devices with various thicknesses of the HAT-CN layers.