Transparent Top-Emitting Organic Light-Emitting Diodes Using a Cs/Al/Ag/ITO Semi-Transparent Cathode

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A top-emitting organic light-emitting diode (TEOLED) was fabricated with a semi-transparent multilayer cathode composed of Cs/Al/Ag and an ITO capping layer. The optical properties of Cs (0.5 nm)/Al (2.0 nm)/Ag (20 nm) at a wavelength of 533 nm exhibited a transmittance of 50 %. The light-output of the TEOELD with the Cs/Al/Ag/ITO showed a maximum luminance of 51,000 cd/m^2 at a current density of 2.32 A/m^2 (corresponding to the forward bias voltage of 11.2 V); the device structure consisted of glass/Ag (100 nm)/ITO (125 nm)/2-TNATA (30 nm)/NPB (15 nm)/Alq$_3$ (55 nm)/Cs (1 nm)/Al (2 nm)/Ag (20 nm)/ITO (100 nm). Also, the external quantum efficiency and the power efficiency at a luminance of 1000 cd/m^2 (corresponding to the forward bias voltage of 6.8 V) were 1.7 % and 2.0 lm/W, respectively. This TEOLED exhibited similar electrical properties, but a little lower light output, compared with a reference TEOLED of the same structure using Alq$_3$ instead of ITO as the capping layer. These characteristics are strongly governed by the thickness, the electrical properties, and optical properties of the semi-transparent multilayer cathode composed of Cs/Al/Ag and by the optical properties of the top capping layer.

I. INTRODUCTION

Organic light-emitting diode (OLED) devices have been steadily developed recently as one of the flat-panel-display (FPD) device technologies most suitable for meeting the demands of information-display applications in the future. Top-emitting (TE) OLED structures coupled with a low temperature poly-silicon (LTPS) thin-film-transistor (TFT) backplane are being taken as the most essential key-element techniques to develop active-matrix (AM) OLED displays [1–3]. The TEOLED is known to provide not only a higher aperture ratio than the general bottom-emitting (BE) one but also a higher display image quality because of its geometrical merit allowing a high pixel resolution [4,5]. In addition, various merits can be obtained from the TEOLED structure, such as easier stacking of a color filter (CF) and a color change medium (CCM) on it, enhancement of the luminous efficiency by phase matching of the top capping layer, and color purity control for the irradiated light.

Meanwhile, if the characteristics of intrinsic top emission are to be kept, both high electrical conductivity and high transmittance, which are two conflicting requirements for a cathode, must be simultaneously satisfied. Therefore, the design and fabrication of a semi-transparent (or transparent) conducting cathode in

the visible range is one of the key technologies for a TEOLED.

One approach for improving the light-output efficiency of the conventional TEOLED [5] is to use low-work-function metals such as the alkali metals (*e.g.*, Li [6, 7] and Cs [8,9]), the alkaline earth metals (Mg [10,11] and Ca [12–14]), and so on. Among these metals, the work function of cesium is the lowest as 2.14 eV [15]. Meanwhile, in recent years, various attempts have been made by several research groups to develop new cathode systems composed of a semi-transparent conducting protecting layer (STCPL)/transparent conducting oxide (TCOs) (*e.g.*, Ag-doped Mg/ITO [10, 11] and Ca/ITO [12,]), a multi-metal cathode (*e.g.*, Ca/Ag [13], Ca/Mg [14], and LiF/Al/Ag [16]), or a metal-free cathode (*e.g.*, CuPc/ITO [6], Li-doped BCP/ITO [5], and Li-doped Bphen/ITO [7]). Here, the devices fabricated by using STCPL/TCOs and multi-metal cathodes generally form a micro-cavity structure while the metal-free cathode system was designed to overcome the micro-cavity structure to obtain a high aperture ratio to the top direction [5,7]. Also, in these cathode systems, the STCPL was used to protect the organic layers from the energetic particles occurring during the deposition of the TCO layer.

In this article, a semi-transparent, conducting, protecting cathode of Cs/Al/Ag is introduced for efficient electron injection from the cathode system into the ad-

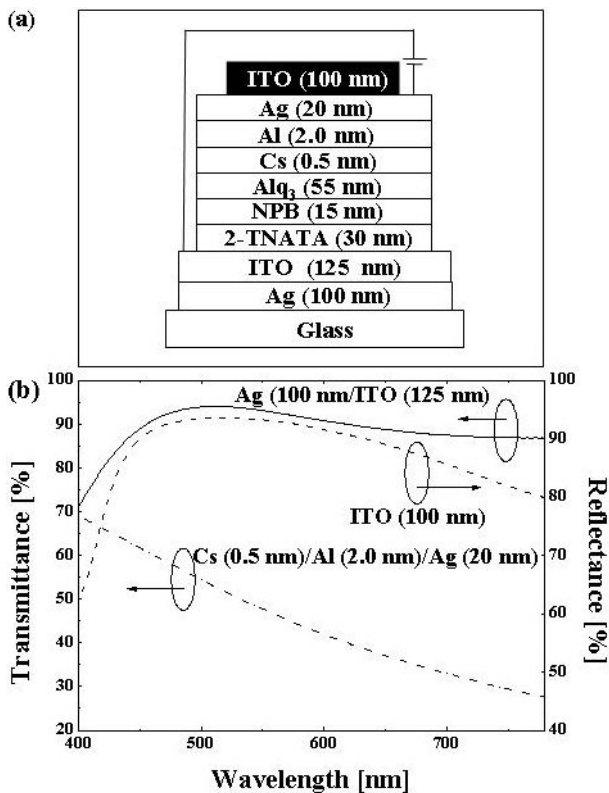*E-mail: gyyeom@skku.edu; Fax: +82-31-299-6565

Fig. 1. (a) Schematic device structure of TEOLEDs consisting of glass/Ag (100 nm)/ITO (125 nm)/2-TNATA (30 nm)/NPB (15 nm)/Alq$_3$ (55 nm)/Cs (x nm)/Al (2 nm)/Ag (20 nm)/ITO (100 nm). (b) Transmittance spectrum of Cs (0.5 nm)/Al (2.0 nm)/Ag (20 nm) (straight line) and ITO (125 nm) (dash-dot line) consisting of a multilayer cathode, and the reflection spectrum of the Ag (100 nm)/ITO (125 nm) anode (dash line).

joining Alq$_3$ organic layer. Also, during the deposition of the capping ITO layer on a glass/anodes/organic layers/Cs/Al/Ag device by dc sputtering, the STCPL plays an important role by the protecting organic layers from high- energy bombardment. For the fabrication of the TEOLED, a suitable damage-free Cs/Al/Ag thickness during the deposition of ITO was also investigated and compared with the TEOLED with a Cs/Al/Ag/Alq$_3$ cathode through the characteristics of the current density - voltage - luminance curve.

II. EXPERIMENTS

A schematic of the device configuration of the TEOLED, which is composed of glass/Ag (100 nm)/ITO (125 nm, about 20 $\sim$ 30 $\Omega/\square$)/2-TNATA (30 nm)/NPB (15 nm)/Alq$_3$ (55 nm)/Cs (0.5 nm)/Al (2 nm)/Ag (20 nm)/ITO (100 nm), is shown in Figure 1(a). The structure of the TEOLED consists of a multilayer anode of Ag/ITO, an organic multilayer of 2-TNATA/NPB/Alq$_3$,

and multilayer cathode Cs/Al/Ag/ITO.

A 100-nm-thick Ag layer consisting of a multilayer anode was vacuum-evaporated on a glass substrate by using a thermal evaporator. Onto this Ag layer, a 125-nm-thick tin-doped indium (ITO) layer was deposited by conventional dc sputtering, followed by a heat treatment; the ITO deposition was carried out in argon at a pressure of 5 mTorr mixed with less than 2 % oxygen at a dc power of 400 W. Sequentially, onto the ITO, an edge-passivation layer of 200-nm-thick silicon oxide (SiO$_2$) was deposited by using electron-beam evaporation to prevent a short-circuit between the top cathode and the bottom anode layers using a shadow mask (not shown in the figure).

Organic layers consisting of 30-nm-thick 4,4',4"-tris[2-naphthylphenyl-1-phenylamino]triphenylamine (2-TNATA) as a hole-injecting layer (HIL), 15-nm-thick 4,4'-bis[N-(1-napthyl)-N-phenyl-amino]-biphenyl (NPB) as a hole-transporting layer (HTL), and 55-nm-thick tris(8-quinolinolato)aluminum (III) (Alq$_3$) as an electron-transporting layer (EIL) were sequentially deposited by using a thermal evaporator system. A 0.5-nm-thick cesium (Cs) layer, a 2.0-nm-thick aluminum (Al) layer, and a 20-nm-thick silver (Ag) layer consisting of a multilayer cathode were deposited onto the multi-organic layers by using a thermal evaporator. Finally, an ITO capping layer was deposited by dc sputtering in argon at a pressure of 5 mTorr mixed with O$_2$ at a power of 100 W. Here, Cs was deposited using a standard SAES getter source. The emissive active area of the devices was 1.4×1.4 mm^2.

Reflectance spectra and transmittance spectra of the electrodes were measured by using a UV-VIS-NIR spectrophotometer with a VW specular wavelength reflectance (Cary 5000 UV/VIS/NIR, Varian Inc.) and a UV spectrophotometer (UV S-2100, SCINCO Inc.), respectively. The resistivity was measured by using a four-point probe (CMT-SERIES, CHANG MIN Co. Ltd.). The current - voltage - luminance characteristics were measured using a source-measure unit (2400, Keithley Instrument Inc.) while the emission intensities from the TEOLEDs devices were measured by using the photocurrent induced on a silicon photodiode (Oriel 71608) with a picoammeter (485, Keithley Instrument Inc.). The electroluminescence (EL) spectra of the as-fabricated devices were measured by using optical emission spectroscopy (PCM-420, SC Tech. Inc.).

III. RESULTS AND DISCUSSION

If a low operating voltage is to be achieved in the TEOLED, it is necessary to have ohmic interfaces between the adjoining organic layer and the charge injecting contacts. The ohmic contact for hole injection can be attained when the HOMO (the highest occupied molecular orbital) of 2-TNATA is same as the work function

(Φ) of an adjoining anode. Also, the ohmic contact for electron injection can be realized by having the LUMO (the lowest unoccupied molecular orbital) energy level of Alq$_3$ the same as the Φ of an adjoining cathode. In our TEOLED structure, the energy barrier (ΔB_h) for hole injection between ITO (Φ: 4.7 eV [6]) and 2-TNATA (IP: 5.1 eV [17]) is 0.4 eV. However, ΔB_e for electron injection between Al (Φ: 4.3 eV [18])/Ag (Φ: 4.3 eV [18]) and Alq$_3$ (EA: 3.1 eV [18]) is about 1.2 eV. Therefore, non-ohmic characteristics generally appear in the current density - voltage curve of OLEDs because ΔB_e for electron injection is far higher than ΔB_h for hole injection. As a function of the thickness, the non-ohmic characteristics of the above cathode/Alq$_3$ structure were proven through the current - voltage curves obtained by Parthasarathy *et al.* [19].

To inject an electron efficiently, the introduction of a low-Φ cesium between an adjoining cathode layer and the Alq$_3$ layer could be expected to reduce the electron injection barrier. Therefore, the use of cesium with a low Φ of 2.14 eV could lead to a device performance with efficient electron injection. Meanwhile, Ag in the Cs/Al/Ag cathode is used as a protecting layer to prevent the oxidation of both Cs and Al, which are sensitive to atmospheric moisture and oxygen. A thin layer of Ag has a relatively low optical absorption and the highest conductivity among all metals. The Ag protecting layer improves the lateral electrical conductance of the cathode as reported by Hung *et al.* [17]. In addition, ITO (refractive index: 1.95 [17]) was used as a high-refractive-index-matching layer for enhancement of the optical transmission through the top of the device and as a semi-passivation layer to protect the device. As the anode of the TEOLED, high reflective mirrors and proper cavities are required to achieve a strong radiative emission as the electric field of states is proportional to the reflectivity of the mirrors and to the reciprocal of the length of the cavity [20]. To obtain a high light-output in this TEOLED study, we used Ag/ITO as a highly reflective anode with a low resistance, and Cs/Al/Ag/ITO was introduced as a highly transmittable cathode with a low resistance.

The TEOLED structure investigated in this study is shown in Figure 1(a). The structure of device 1 was composed of glass/Ag (100 nm)/ITO (125 nm)/2-TNATA (30 nm)/NPB (15 nm)/Alq$_3$ (55 nm)/Cs (0.5 nm)/Al (2 nm)/Ag (20 nm)/ITO (100 nm). Device 2 as the reference has the same structure as device 1 except for device 2 having a 52-nm-thick Alq$_3$ capping layer instead of a 100-nm-thick ITO capping layer. Figure 1(b) shows the respective transmittance spectrum of both Cs (0.5 nm)/Al (2.0 nm)/Ag (20 nm) and ITO (100 nm) consisting of the multilayer cathode as functions of wavelength over the entire visible range. As the figure shown, transparencies of 50 % and 94 % at a wavelength of 533 nm could be obtained, respectively. The reflectance spectrum of the Ag (100 nm)/ITO (125 nm) anode is also shown in Figure 1(b) and shows a reflectance of 94 % at

a wavelength of 533 nm.

The low transmittance of Cs (0.5 nm)/Al (2.0 nm)/Ag (20 nm) is attributed to the thickness of silver being thicker than the skin depth (δ) of silver. δ of silver can be expressed as follow: $\delta = \lambda/4\pi n_i$ (λ: wavelength, n_i: refractive index). Taking $\lambda = 5 \times 10^{-7}$ m and $n_i = 3$, we obtain a δ of approximately 13 nm [21]. Although 20-nm-thick silver is thicker than the skin depth, it was applied to the thickness for efficient electrical conduction. Meanwhile, in the visible range, as the optical wavelength is decreased, the transmittance of the Cs (0.5 nm)/Al (2 nm)/Ag (20 nm) cathode is relatively increased while the reflectance is decreased. Therefore, this multilayer cathode is more suitable for application to the TEOLED. In addition, Cs (0.5 nm)/Al (2 nm)/Ag (20 nm) and ITO (100 nm) composing the multilayer cathode showed resistivities of 8.0×10^{-6} and 9.8×10^{-4} $\Omega\cdot$cm, respectively, while the Ag/ITO multilayer anode showed a resistivity of about 4.0×10^{-6} $\Omega\cdot$cm.

Using the TEOLED with the cathode system and the anode system (device 1), we measured the current - voltage - luminance characteristics of the device, and the results are shown in Figure 2 for (a) the current density and (b) the luminance as functions of the forward bias voltage. The current density - voltage - luminance characteristics are also summarized in Table 1. In Table 1, the characteristics of the TEOLED device fabricated with a 52-nm-thick Alq$_3$ capping layer instead of a 100-nm-thick ITO capping layer (device 2) are presented as a reference for comparison. As Figure 2 and Table 1 shown, at a luminance of about 1000 cd/m^2 (L_{1000}), the current densities of device 1 and device 2 were 25.9 mA/cm^2 (6.8 V) and 24.3 mA/cm^2 (6.6 V), respectively.

The inset of Figure 2(b) shows the EL spectrum for device 1 measured at the normal viewing angle and at L_{1000}. As in the inset of Figure 2(b) shown, the maximum EL peak was found at a wavelength of 533 nm. The maximum EL peak of 533 nm was obtained by adjusting the macro-cavity effect [19], which could be controlled by changing the thickness of ITO composing the multilayer anode.

The maximum luminances (L_{max}) for devices 1 and 2 were 56000 and 77200 cd/m^2, respectively, as shown in Table 1 and Figure 2(b). Therefore, device 2 with the Alq$_3$ top capping layer showed a slightly higher luminance than device 1 with the ITO capping layer. The slightly higher luminance for device 2 appears to be from refractive-index matching with air when the emitted light is transmitted out of the top capping layer. This could be decided by Δn, which is the difference in the refractive index between the top capping layer and air. Device 2 with a capping layer of Alq$_3$ (n: 1.7) has a Δn of 0.7, but device 1 with a capping layer of ITO (n: 1.95) has a Δn of 0.95. Therefore, a higher out-coupling property could be obtained for device 2 due to the smaller Δn. However, the electron injection properties of two devices are similar because the electrical properties of the multilayer cathode commonly depend on the STCPL, that

Table 1. Current density - voltage - luminance characteristics for devices 1 and 2.

Devices	Capping cathode layer	η_{ext} (%)	η_{PE} (lm/W)	V_{1000} (V)	V_T (V)	L_{max} (cd/m^2)
Device 1	ITO (125 nm)	1.7	2.0	6.8	2.8	56,000 (11.2 V)
Device 2	Alq$_3$ (52 nm)	1.9	2.4	6.6	2.8	77,200 (11.2 V)

η_{ext}, η_{PE}, and V_{1000} are values at 1,000 cd/m^2, respectively. V_T and L_{max} are the turn-on voltage at a luminance of 0.1 cd/m^2 and the maximum luminance, respectively.
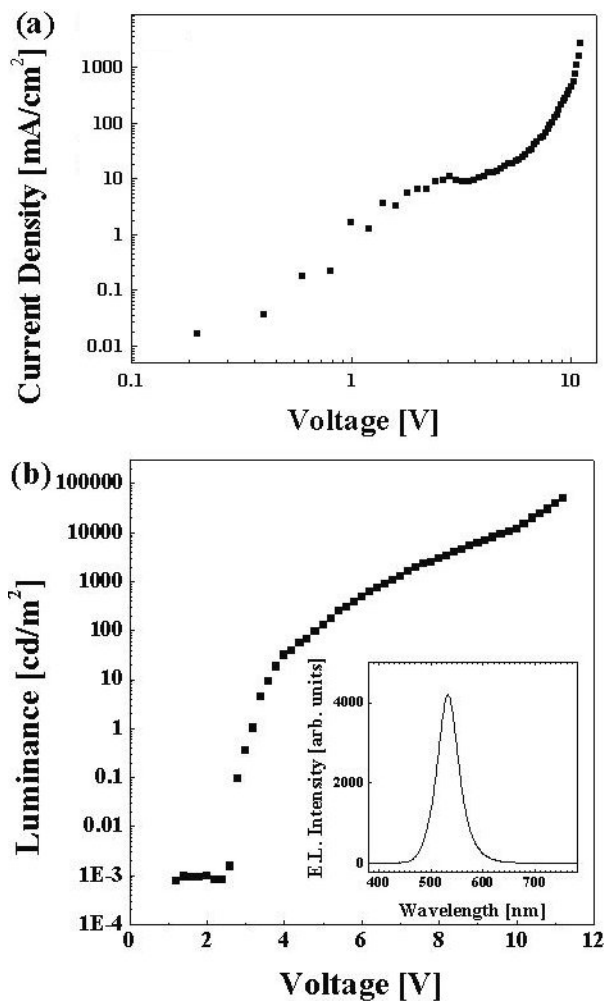

Fig. 2. (a) Current density as a function of voltage for the TEOLED composed of glass/Ag (100 nm)/ITO (125 nm)/2-TNATA (30 nm)/NPB (15 nm)/Alq$_3$ (55 nm)/Cs (0.5 nm)/Al (2.0 nm)/Ag (20 nm)/ITO (100 nm). (b) Luminance as a function of voltage for the same device. The inset is the electroluminescent spectrum at a luminance of 1000 cd/m^2.
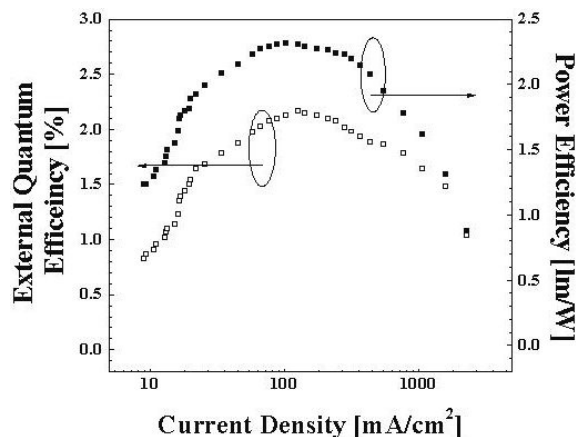

Fig. 3. External quantum efficiency - current density characteristics [open squares] and power efficiency - current density characteristics [closed squares] of the TEOLED having an ITO capping layer.

is, on the Cs/Al/Ag layer and, especially, on the energy barrier differences of Cs and Alq$_3$. Also, the turn-on voltages of devices 1 and 2 were the same, 2.8 V.

During the deposition of ITO onto glass/Ag/ITO/ organic layers/Cs/Al/Ag, if the thickness of Cs/Al/Ag cathode system was not thick enough, the device could be damaged by the plasma through oxidation and phys-

ical damage of the organic layers. In fact, the TEOLED with the Cs (0.5 nm)/Al (2.0 nm)/Ag (below 15 nm) cathode showed a poor device performance after sputter deposition of the 100-nm-thick ITO capping layer (not shown). However, the similar electrical characteristics between devices 1 and 2 obtained in this study show that the cathode system composed of Cs (0.5 nm)/Al (2.0 nm)/Ag (20 nm) is sufficiently not to allow the damage by the plasma during the ITO deposition.

Figure 3 shows the external quantum efficiency (η_{ext}) and the power efficiency (η_{PE}) for device 1 as functions of the current density, and the results are summarized in Table 1, together with the results for device 2. As Figure 3 and Table 1 shown, η_{ext} at L_{1000} for devices 1 and 2 were 1.7 and 1.9 %, respectively, and η_{PE} at L_{1000} were 2.0 and 2.4 lm/W, respectively. Therefore, device 2 showed a slightly higher η_{PE} and a slightly higher η_{ext}. Although the TEOLED with the Cs/Al/Ag/ITO cathode showed slightly worse optical properties than the TEOLED having the Cs/Al/Ag/Alq$_3$ cathode, it is believed that the TEOLED with the ITO capping layer was not damaged during the deposition of ITO capping layer due to the suitably thick Cs/Al/Ag layer, and a highly efficient TEOLED could be obtained due to the low energy barrier between the low Φ of Cs and the electron affinity of Alq$_3$.

IV. CONCLUSIONS

A TEOLED using Cs/Al/Ag as the cathode layer and ITO as the capping layer was successfully fabricated by sequential deposition. The multilayer cathode consisting of Cs (0.5 nm)/Al (2.0 nm)/Ag (20 nm) showed a transmittance of 50 % and the Ag (100 nm)/ITO (125 nm) anode showed a reflectance of 94 % at a wavelength of 533 nm. When the current density - voltage - luminance characteristics of the TEOLED with the ITO capping layer were compared with those of the TEOLED with the Alq_3 capping layer, the electrical properties of the TEOLEDs were similar to each other, even though the optical properties were a little better for the TEOLED with the Alq_3 capping layer. Changes in the electrical properties do not show in the current density - voltage curve of the TEOLEDs after plasma exposure during the ITO deposition. The multilayer cathode (Cs/Al/Ag) is thick enough, not to allow damage from the plasma during the deposition of the ITO capping layer. Even though the TEOLED with the ITO capping layer showed a slightly lower optical coupling, compared to the TEOELD with the Alq_3 capping layer, due to the refractive index differences between the capping layer and air, the optical characteristics of the TEOLED with the ITO capping layer were still excellent. The light-output of this TEOELD exhibited a L_{max} of 56000 cd/m^2 at a current density of 2.32 A/m^2 (corresponding to a forward bias voltage of 11.2 V). Also, the values of η_{ext} and η_{PE} at L_{1000} (about 6.8 V) were 1.7 % and 2.0 lm/W, respectively. Thus, a successful TEOLED was possible due to the small energy barrier between the Cs and the Alq_3, which cause higher electron injection from the Alq_3 to the Cs with a low Φ.

ACKNOWLEDGMENTS

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