Transparent organic light-emitting devices with CsCl capping layers on semitransparent Ca/Ag cathodes

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Abstract

We have developed transparent organic light-emitting devices (TOLEDs) with CsCl capping layers deposited on top of semitransparent Ca/Ag cathodes. The CsCl layer was deposited by the thermal evaporation method which does not result in any damage to the underlying organic layers. The transmittance was enhanced by depositing the CsCl layer on the Ca/Ag cathode. The current efficiency measured at the cathode side increased by the enhanced transmittance of the cathode, whereas the anode-side current efficiency was determined by the reflectance of the cathode. In a TOLED with semitransparent Ca/Ag/CsCl layer, the microcavity effect was not profound so that the electroluminescence spectrum was not seriously changed by the CsCl capping layer.

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1. Introduction

Organic light-emitting devices (OLEDs) have been attracted much attention in the past two decades because they have several advantages such as wide viewing angle, fast response, low-voltage operation, thinness, and two-dimensional emissions in display and light applications [1,2]. OLEDs are composed of the functional organic layers between transparent anode and reflective cathode. Since the organic layers are optically transparent in the visible range, OLED becomes transparent when the reflective cathode is replaced with the transparent one. These transparent OLEDs (TOLEDs) have been extensively researched because they can provide wide applications such as car windshield, architectural window, and eyewear [3–5].

As a transparent cathode, sputtered indium tin oxide (ITO) has been suggested because of its high optical transparency and low resistivity. However, organic layers are damaged by energetic particles during the deposition of ITO. Although the several approaches, such as extremely low rate of deposition (∼0.005 nm/s) [6], facing target sputtering [7], and organic buffer layer [4], have been tried to reduce the sputter-induced damage, it is still difficult to avoid the deterioration of device performances. Since the first report on the thermally evaporated ultrathin LiF/Al cathode which does not damage the underlying organic stacks [8], many researchers have suggested the thermally evaporated damage-free semitransparent metal cathodes such as LiF/Al/Ag [9], Sm/Au [10], and Ba/Ag [11]. Pode et al. reported the Ca/Ag cathode which has a high transmittance and a low sheet resistance [12–14]. Furthermore, it has been reported that the transmittance of the semitransparent metal cathode can be further improved by depositing organic or inorganic capping layer on top of the cathode [8,15].

In top emission OLED (TEOLED) where the generated light in the emission layer is strongly reflected by the reflective metal anode, the effect of transmittance of semitransparent cathode on the device performance is controversial. Riel et al. reported that the performance of TEOLED is dominated by the microcavity effect rather than the transmittance of cathode [16]. On the other hand, we found that the transmittance is strongly related to the device performance in our TEOLED [15]. We increased the efficiency of the top emission device using CsCl capping layer on the semitransparent Ca/Ag cathode [15]. However, TOLED is different from TEOLED because the light is not strongly reflected by the anode in the transparent device since the transparent anode is used instead of the reflective metal anode. Furthermore, in transparent device structure, the device performance may be affected by the reflectance of the semitransparent metal cathode since the reflected light by the cathode may be extracted out of the transparent anode side. In this paper, we demonstrate the effect of transmittance and reflectance of the semitransparent Ca/Ag cathode with CsCl capping layer on the cathode- and anode-side performances of the TOLED. We have studied the effect of CsCl layer on the transmittance and reflectance of the Ca/Ag cathode. We have also investigated the effect of CsCl capping layer on the current conduction, current efficiency, and emission characteristics of the TOLEDs with the semitranspar-
ent Ca/Ag cathode. We demonstrate that the current efficiency is strongly related with the transmittance and reflectance of the cathode.

2. Experimental details

In order to investigate the effect of CsCl layer on the optical transmittance and reflectance of the cathode, CsCl layers were prepared on the Ca/Ag coated glass substrates. Ca (10 nm) and Ag (10 nm) layers were sequentially deposited on the cleaned substrates, followed by the deposition of CsCl layers without breaking a vacuum. All the layers were deposited by a thermal evaporation method with a deposition rate of 2–3 Å/s in a background pressure of $1 \times 10^{-6}$ Torr. The thickness of the CsCl layer was varied from 0 to 200 nm. The transmittance and reflectance were measured using a JASCO V-560 spectrophotometer with a normal incidence of monochromatic light at the sample surface.

In order to investigate the effect of CsCl capping layer on the device performances, the TOLEDs were fabricated on the ITO coated glass substrates. The sheet resistance of the ITO film was about 10 $\Omega/\square$. After defining ITO anode patterns using a standard photolithography process, the substrates were cleaned with isopropyl alcohol and deionized water, followed by exposing to oxygen plasma at 150 W for 5 min. All the organic, metal, and CsCl layers were deposited by using a thermal evaporation method in a base pressure of $1 \times 10^{-6}$ Torr. A 15 nm thick 4,4′,4″-tris[N-(2-naphthyl)-N-phenyl-amino]triphenylamine (2-TNATA) layer was deposited on the patterned ITO substrate, followed by the deposition of a 35 nm thick 4,4′-bis-[N-(1-naphthyl)-N-phenyl-amino]diphenyl (α-NPD) layer. Then, a 35 nm thick tris-(8-hydroxyquinoline) aluminum (Alq3) layer was co-deposited with 1 wt.% fluorescent dopant, 10-(2-benzothiazolyl)-1,1′,7,7′-tetramethyl-2,3,6,7-tetrahydro-1H,5H,11H-[1]benzopyrropropyra-nol[6,7,8-i]quinolinilizin-11-one (C545T). Next, a 5 nm thick 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) layer was evaporated, followed by the deposition of a 5 nm thick Alq3 layer. After depositing organic layers, a 10 nm thick Ca and a 10 nm thick Ag layer were sequentially evaporated through a shadow mask. Then, various thicknesses of CsCl capping layers were deposited on the Ca/Ag cathodes of TOLEDs. The completed device structure is ITO/2-TNATA (15 nm)/Ag (10 nm)/Ca (10 nm)/Ag (10 nm)/CsCl (0–200 nm). All the fabricated devices were encapsulated using transparent glass caps without exposing to air in a nitrogen glove box. Current density–voltage–luminance characteristics of the devices were measured using computer controlled Keithley 2400 source-measure units and a calibrated fast silicon photodiode (FDS010). Electroluminescence (EL) spectra were measured with a spectroradiometer (Minolta CS1000).

3. Results and discussion

In order to investigate the effect of CsCl layer on the transmittance and reflectance of the cathode, various thicknesses of CsCl layers were deposited on the Ca/Ag coated glass substrates. Fig. 1 shows the transmittance curves as a function of wavelength for the Ca/Ag/CsCl layers. The CsCl layer thickness was varied from 0, 60, 75, 100, and 150 nm. The transmittance exhibited a maximum transmittance of 93% at a wavelength of 450 nm. Since the emission peak of fluorescent green dopant, C545T, is located at 520 nm, the transmittance was measured at this wavelength. The Ca/Ag layer exhibits a transmittance of 69% at 520 nm. The transmittance increases to 91% when the 60 nm thick CsCl layer is deposited on the Ca/Ag layer. However, the transmittance becomes 67% when the CsCl thickness is 150 nm. These results indicate that the CsCl layer (refractive index = 1.4) strongly modifies the transmittance of the Ca/Ag/CsCl layers. The CsCl layer thickness increases to 150 nm at a wavelength of 520 nm. On the other hand, the reflectance decreases by depositing a 60 nm thick CsCl layer, and then it increases with increasing CsCl thickness. This modification of reflectance is consistent with the transmittance change by the CsCl layer.

In order to investigate the effect of CsCl capping layer on the device performances, the TOLEDs with a structure of ITO/2-TNATA (15 nm)/α-NPD (35 nm)/Alq3/C545T (1%, 35 nm)/BCP (5 nm)/Alq3 (5 nm)/Ca (10 nm)/Ag (10 nm)/CsCl (x nm) were fabricated. Fig. 2 shows the reflectance spectra of the fabricated TOLEDs. The reflectance spectra of the TOLEDs exhibit almost similar phenomena with those of Ca/Ag/CsCl layers, except the drop of transmittance in the short wavelength region resulting from absorption of light by the organic layers in the devices. The TOLED with a semitransparent Ca/Ag cathode exhibits a transmittance of 58% at a wavelength of 520 nm. On the other hand, a 60 nm thick CsCl layer on the cathode results in a transmittance of 72%. However, the transmittance decreases to 50% as the CsCl thickness increases to 150 nm. Therefore, the device transmittance increases with increasing the cathode transmittance.

While the CsCl layer affects the device transmittance, the current conduction characteristic of the device is not altered by the CsCl capping layer, as shown in the current density–voltage curves.
Fig. 3. Transmission spectra for the TOLEDs with different thickness of CsCl capping layers.

The devices exhibit almost same current density–voltage characteristics despite of the various thicknesses of the CsCl layers. For example, the device with Ca/Ag cathode requires an applied voltage of 8.0 V for a current density of 14.1 mA/cm². The operating voltages are almost same in the TOLEDs with 60–150 nm thick CsCl layers. These results indicate that the underlying organic stacks are not damaged by the CsCl capping layer. It is due to the damage-free nature of the thermal evaporation.

Fig. 5 shows the luminance–voltage and current efficiency–current density curves measured at the cathode side for the TOLEDs. The luminance and current efficiency are dependent on the CsCl capping layer. For example, a luminance of 7140 cd/m² is achieved in the TOLED with Ca/Ag cathode. On the other hand, the device with a 60 nm thick CsCl capping layer exhibits a higher luminance of 11100 cd/m² at the same voltage. This increase of luminance at the same voltage is caused by an improvement of current efficiency, as shown in Fig. 5(b). The current efficiency of the device with a 60 nm thick CsCl capping layer is about 2 times better than the device without CsCl layer. However, the current efficiency decreases in the thicker CsCl device, although the efficiency is still higher than the Ca/Ag device. Therefore, the cathode-side current efficiency shows the same tendency with the transmittance of the Ca/Ag/CsCl layer. The higher cathode transmittance results in a higher cathode-side current efficiency. In our device the higher cathode transmittance means that the light generated in the device can be more effectively extracted out of the cathode.

It should be noted that the luminance and current efficiency measured at the anode side exhibit different tendency from the cathode side. Fig. 6 shows the luminance–voltage and current efficiency–current density curves measured at anode side of the TOLEDs. The luminance and current efficiency curves exhibit different dependences on the CsCl thickness from the cathode side. The TOLED without CsCl capping layer exhibits a luminance of 31400 cd/m² at 14 V and a current efficiency of 7.7 cd/A. The coating
of CsCl capping layer on the Ca/Ag cathode results in an increase of current efficiency as shown in Fig. 6(b). The TOLED with Ca/Ag/CsCl (60 nm) exhibits an anode-side current efficiency of 8.4 cd/A. As the CsCl thickness increases to 150 nm, the current efficiency further increases to 11 cd/A. Hence, the device with a 150 nm thick CsCl exhibits 140% higher current efficiency than the device without CsCl layer. This increase in current efficiency is related to the reflectance of the cathode. As shown in Fig. 2, the reflectance of the cathode is higher in the 150 nm thick CsCl layer. This higher reflectance means that more photons are reflected by the cathode, being extracted out of the anode side. The transmittance and reflectance of the cathode also affect the ratio between the current efficiencies measured at the anode and cathode sides. Fig. 7 shows the anode- and cathode-side current efficiencies as a function of CsCl thickness. The anode-side current efficiency is higher than the cathode-side one. It is due to the reflection of photons by the cathode. Despite the cathode is semitransparent, the cathode is still reflective compared to the ITO anode. Hence, the more photons are exiting out of the anode side rather than cathode.

Fig. 8 shows the EL spectra measured at the anode and cathode side for the TOLEDs. The spectrum exhibits a peak intensity at 515–520 nm, which corresponds to the emission from green fluorescent dopants, C545T molecules. The EL spectrum is not seriously changed by the CsCl capping layer. Therefore, the figure suggests that the microcavity effect is weak in our TOLEDs. Although the reflection by the semitransparent cathode affects to the anode-side luminance and current efficiency, the multiple interferences resulting in strong modification of emission spectrum is not profound in our devices.

4. Conclusions

We have investigated the effect of CsCl capping layer deposited on the semitransparent Ca/Ag cathode of the TOLED. By using a CsCl capping layer, the transmittance of the cathode can be increased by 93%. While the electrical conduction characteristics of the device are not changed by the capping layer, the cathode-side and anode-side current efficiencies can be improved by 2 and 1.4 times, respectively, by depositing CsCl capping layer on the Ca/Ag cathode. These improvements of cathode-side and anode-side current efficiencies are attributed to the enhancements of transmittance and reflectance by the CsCl capping layer.

References