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# Enhancement in light extraction efficiency of organic light emitting diodes using double-layered transparent conducting oxide structure

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ABSTRACT

We examined double-layered transparent conducting oxide (TCO) anode structures consisted of zinc-doped indium oxide (IZO) over the gallium-doped zinc oxide (GZO), and IZO over the GZO with electrochemical treatment. In bottom type OLEDs, power efficiency and current efficiency were enhanced by a factor of 1.50 and 1.14 at a current density of 10 mA/cm$^2$ in IZO/GZO anode structure, compared to the only IZO anode structure. Due to the reduced sheet resistance of the IZO/GZO TCO surface, the operating voltage of the OLED with IZO/GZO anode structure was lowered, leading to mostly enhance power efficiency. More enhanced in power efficiency and current efficiency by a factor of 1.21 and 1.25 at a current density of 10 mA/cm$^2$ were achieved in IZO/GZO anode structure with electrochemical treatment, compared to the IZO/GZO anode structure due to the change of the surface morphology of the GZO and the existence of the nanoporous layer beneath the GZO surface by an electrochemical treatment. In total, double-layered IZO/GZO anode structure with electrochemical treatment was revealed at an enhancement factor of 1.80 in power efficiency and 1.42 in current efficiency, compared to the only IZO anode structure.


## 1. Introduction

Organic light emitting diodes (OLEDs) have been a topic of extensive research for several decades and have been commercialized recently in devices such as small mobile displays and larger displays for OLED televisions. Another application area for OLEDs is in solid-state lighting, which is on the verge of being commercialized for use in next-generation lighting applications [1]. One of the main challenges of advanced OLED technology is attaining higher power efficiency and current efficiency, which are enhanced light extraction efficiency [2,3]. However, significant loss of light within the OLED occurs as the following reasons. In the case of bottom-type OLEDs, a large amount of the light emitted from the organic materials can be trapped within the organic layer and the transparent conducting oxide (TCO) layer because of a wave-guiding phenomenon, referred to as the internal modes, and within the substrate layer because of total internal reflection, referred to as the substrate modes [2–11]. Another form of loss occurs because of "absorption" on the metal cathode adjacent to the electron transport layer (ETL), called the surface plasmon-polariton (SPP) loss [3,12–14]. Based on ray

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optics by our simulation calculation, the light loss in the OLEDs is estimated to be about 45% in internal modes, about 20% in substrate modes, and about 10% in SPP loss, and finally, about 25% of the light is out-coupled from the substrate. To extract the light trapped within the substrate, film-type light-extraction layers with some periodic patterns have been attached to the outer substrate [15,16]. There is a higher potential for light extraction efficiency due to the highest loss percentage of the light in internal modes, and this leads to challenges in developing more-power-efficient OLED lighting. Several efforts have been made to enhance the internal light-extraction efficiency, for example, by inserting alternative layers with different refractive indices between the TCO electrode and the glass substrates [17], by patterning the TCO surface or planarization process [18], by stamping a buckled structure between the TCO and the glass substrate [19], and by creating a regular low-index-grid pattern on top of the TCO [20]. Although these results showed great improvement in light extraction efficiency, they suggested that there is still room for maximizing the extraction of the wave-guided light in OLEDs.

In this study, we present a new approach to enhance the light extraction efficiency in bottom-type OLEDs using a double-layered TCO anode structure. A double-layered TCO was consisted of a relatively high-refractive index (RI) zinc-doped indium oxide (IZO) layer over the relatively low-RI gallium-doped zinc oxide (GZO) layer. Additionally, electrochemical treatment process could change the surface morphology of the GZO layer and generate nanopores structures within the surface region of the GZO layer. The double-layered TCO structure with electrochemical process played an important role in the enhancement of the light extraction efficiency, compared to the IZO anode-based OLEDs.

## 2. Experimental

GZO film with a thickness of 700 nm was uniformly grown on an Eagle XG (Corning) glass substrate that was 150 mm × 150 mm in size, by atmospheric pressure chemical vapor deposition (APCVD). The APCVD precursors were modified diethyl-zinc ($Zn(C_2H_5)_2$) and trimethyl gallium ($C_3H_9Ga$), which were used to prevent the flammability of the system. The glass substrate temperatures were maintained at approximately 500 °C. A detailed description of the APCVD system was published elsewhere [21]. In order to generate nanoporous structures on the GZO surface, it was immersed in ethylene glycol containing 0.5 wt% $NH_4F$ and was electrochemically treated for 100 s (input voltage and current were 20 V and 0.7 A, respectively) [22]. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were used for observing the surface morphologies and performing structural analysis. IZO with a thickness of 200 nm using a sputtering system (the sputtering target contains 10 wt% of zinc and 90 wt% of indium oxide) was deposited on glass, GZO, and nanoporous GZO (np-GZO) to form the anodes of the bottom type OLEDs. The optical transmittance and the sheet resistance of the IZO/glass, IZO/GZO/glass, and IZO/np-GZO/glass were analyzed using a UV–visible spectrophotometer and a four-point probe system, respectively. For evaluating the OLEDs performances, OLEDs with anodes comprising of IZO, IZO/GZO, and IZO/np-GZO were fabricated using green fluorescent materials. IZO/glass, IZO/GZO/glass, and IZO/np-GZO/glass were simultaneously cleaned with isopropyl alcohol (30 min) by ultrasonic cleaning and dried for 30 min at 100 °C. To eliminate organic residues and to increase the work function on the anode surface, UV/ozone treatment was performed on all samples. Vacuum deposition of a 180 nm-thick hole injection layer (HIL)/hole transport layer (HTL), 40 nm-thick green emitting layer, 25 nm-thick electron transport layer (ETL) and a 1 nm-thick electron injection layer (EIL) was performed in sequence at a base pressure of $\sim 2 \times 10^{-7}$ Torr in an organic chamber. Subsequently, the samples were transferred to a metal chamber and a 120 nm-thick aluminum cathode layer was deposited at a base pressure of $\sim 2 \times 10^{-7}$ Torr. Finally, all the samples were encapsulated in an $N_2$ glove box. Luminance-current density–voltage source meter (Keithley 2400) was employed for the current/voltage and luminance/voltage measurements.

## 3. Results and discussion

Fig. 1a shows a side view of the GZO on glass substrate grown by APCVD with a thickness of ~700 nm. The change in morphology near the top surface of the GZO by the electrochemical treatment is shown in Fig. 1b. The rough surface of the GZO was changed into a surface with triangular protrusions with a height of around 100 nm. $F^-$ ions from the buffered neutral electrolytes containing $NH_4F$ are likely to etch the GZO surface, and the etching rate along the GZO grain boundaries is faster than that of the GZO grains, generating the triangle-shaped surface. Additional analysis was done using TEM and the images are shown in Fig. 2. A distinct difference was observed between the electrochemically treated GZO and the as-grown GZO, as shown in Fig. 2a and b. Around 100 nm-thick, visually distinguishable layer was generated beneath the GZO surface by the electrochemical treatment. The presence of this layer within the electrochemically treated GZO is attributed to the distribution of nano-voids. We observed that the structures of the regions (marked with the dotted line) are different, as seen from the selected area electron diffraction patterns (SAED) (the insets in Fig. 2c and d). The long-range order in GZO is likely to be deteriorated by the electrochemical treatment. A detailed structural analysis of this phenomenon is currently under progress. Other regions, such as the middle and bottom regions in the GZO layer showed no distinct change after the electrochemical treatment (not shown here).

The results of sheet resistance measurement for the IZO/glass, IZO/GZO/glass, and IZO/np-GZO/glass are shown in Table 1. The IZO/GZO/glass and the IZO/np-GZO/glass structures have a sheet resistance of 8.6 and 9.9 Ω/sq, which is much lower than the IZO/glass structure because of the thick double-layered TCO structure. Meanwhile, the sheet resistance of the IZO/np-GZO/glass was a little higher