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Electrical and Optical Properties of IZTO Thin Film for OLED Anode

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Indium Zinc Tin Oxide (IZTO) thin-films for OLED (organic light-emitting diode) anodes were prepared through a Facing Target Sputtering (FTS) system under various sputtering conditions. The FTS system has several advantages such as hetero-sputtering, low working pressure, and high plasma density. When two sheets of targets are installed on an FTS system, one of the targets is IZO (In_2O_3 90wt%, ZnO 10 wt%) and the other target is ITO (In_2O_3 90wt%, SnO_2 10wt%). As-deposited IZTO thin-films were investigated by a UV/VIS spectrometer, an X-ray diffractometer (XRD), a Hall-Effect measurement system, and an atomic force microscope (AFM). The properties of the OLEDs were measured through a J-I-V measurement system.

The IZTO thin-films that were deposited on glass substrate showed an average transmittance of over 80% in a visible range except for IZTO thin-film that was deposited under an O_2 gas flow rate of 0.1 [sccm].

Keywords: OLED; FTS; IZTO; hetero-target

Introduction

Transparent conducting oxides (TCO) have been comprehensively used in optoelectronic devices, such as transparent thin-film transistors (TTFTs) and transparent electrodes in flat-panel displays, flexible displays, and solar cells [1,2]. Generally, it is well-known that the optical, electrical, and mechanical properties, microstructure, and surface uniformity are very important factors for TCO films for display applications [3]. Among many TCO materials, ITO (Indium Tin Oxide) thin-films must be deposited at a temperature higher than 250°C or annealed at a temperature higher than 300°C to have high electrical conductivity and high transmittance [4]. This high-temperature processing makes the ITO films rough due to crystallization, which leads to a significant deterioration in the reliability of the concerned device. For this reason, in recent years, there have been many efforts to replace ITO thin-films by amorphous IZTO (α -IZTO) films. α -IZTO thin-films have several advantages over ITO thin-films; these are now described. α -IZTO thin-films have higher structural stability at higher temperatures than ITO thin-films. The amorphous structure of IZTO thin-films is maintained up to a temperature as high as 350°C, which ensures good stability in both electrical and optical properties. α -IZTO thin-films have higher chemical stability than

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ITO thin-films. In particular, for FPD applications, the TCO films must be prepared at low temperatures below 200°C to avoid thermal stress on the TFT devices or the flexible substrates. However, ITO thin-films that are deposited at lower temperatures have lower resistance to moist heat, higher electrical resistivity, lower optical transmittance, and poorer chemical stability. Besides, α -IZTO thin-films have better wet-etch performance [4–6]. In addition, α -IZTO thin-films adapt to OLED (organic light-emitting diode) applications because α -IZTO thin-films have been reported to have a higher work function than ITO thin-films [7,8].

In this work, IZTO thin-films for OLED anodes were prepared by a Facing-Target Sputtering (FTS) system. In the selection of a material for an OLED anode, important considerations are the need for low resistivity ($>10^{-4} \Omega\text{-cm}$), high work function, smooth surface, and high transmittance ($>80\%$). In order to obtain an IZTO thin-film with these properties towards use in OLED anodes, we prepared IZTO thin-film under various sputtering conditions, which were functions of the O_2 gas flow rate and the input power at room temperature. The effects of the sputtering conditions have been investigated in detail.

Experimental

In the case of a general sputtering system, the position of the substrate for depositing the thin film occurs in the space where the substrate is exposed to plasma. Therefore, there is a limit to the production of thin films of high quality because such film is inevitably damaged through bombardment by high-energy particles. However, the facing-target sputtering (FTS) system can restrain the bombardment by particles that have high energy because the substrate is positioned away from the plasma. In addition, γ -electrons can engage in reciprocal motion between the two targets that face with each other owing to the cathode electric potential of the two targets and raise the ionization rate for the working gas. Thus, the FTS can maintain a stable discharge condition at low working pressure so that high-quality thin-film can be deposited. Figure 1 shows the schematic diagram of the FTS system [9,10]. The FTS system comprises two targets that face each other: IZO (In_2O_3 90 wt%, ZnO 10 wt%, 2-inch diameter) and ITO (In_2O_3 90 wt%, SnO_2 10 wt%, 2-inch diameter). The spacing between the substrate and the targets was fixed at 90 mm and the spacing between the facing targets was fixed at 50 mm. The chamber was evacuated to a pressure of 6×10^{-7} Torr.

Before the deposition, the substrate was ultrasonically cleaned in deionized water-isopropyl alcohol (IPA) and subsequently dried in flowing N_2 gas. The sputtering conditions for the preparation of IZTO thin-films are given in Table 1. The electrical properties were measured by a Hall-Effect measurement system (ECOPIA, HMS-3000). The structural and optical properties were measured by an X-ray diffractometer (Rigaku, D/MAX-2200), an atomic force microscope (PSIA, XE-150), and a UV/vis spectrometer (HP8453, Hewlett-Packard). To examine the electrical and optical properties of OLEDs that were fabricated on α -IZTO thin-films, the OLEDs were prepared on plasma-treated IZTO thin-film and on non-plasma-treated IZTO thin-film through a thermal evaporation system. We used N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD 40nm, TCI), tris(8-hydroxyquinolinato)aluminum (Alq_3 60nm, TCI), Lithium fluoride (LiF 1nm, Aldrich), and Aluminum (Al 50nm, Aldrich) as the hole transport layer (HTL), the emitting layer (EML), the electron injection layer (EIL), and the cathode layer, respectively. The current density-voltage-luminance (J-V-L) characteristics of the fabricated OLEDs were measured with a Keithley 2400 device and a luminance meter.

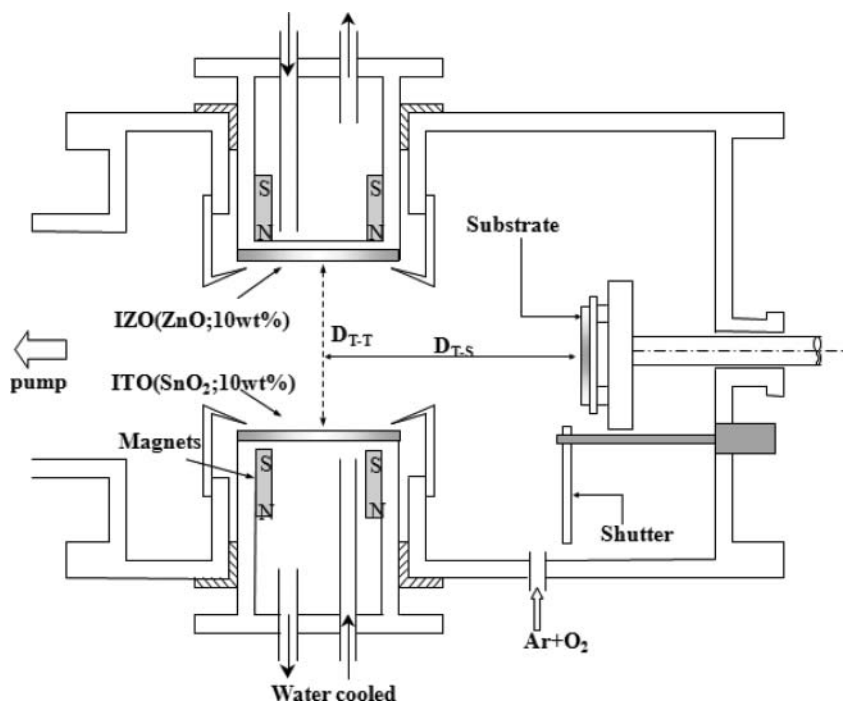


Figure 1. Diagram of facing targets sputtering system

Results and Discussion

Figure 2 shows the electrical properties of the deposited IZTO thin-films as a function of the input power and the O₂ gas flow rate. As a result, when the IZTO thin-films were deposited at an input power of 45 W, we could obtain the highest mobility and the lowest resistivity for the OLED anodes, as shown in Fig. 2. The mobility increased with the O₂ gas flow rate, which is due to the decrease in the ionized scattering centers of the carrier sights, such as electrically active oxygen vacancies [11]. When IZTO thin-film was prepared under an O₂ gas flow rate of 1.1 sccm, it exhibited the highest mobility of 51.17 [cm²/V·s]. The carrier

Table 1. Sputtering conditions

Deposition parameter	Conditions
Target	IZTO(ITO: In ₂ O ₃ 90wt%, SnO ₂ 10wt%, IZO: In ₂ O ₃ 90wt%, ZnO 10wt%)
D _{T-S}	90mm
D _{T-T}	50mm
Substrate	Glass
Base pressure	2.67 × 10 ⁻⁴ Pa
Working pressure	0.133 Pa
O ₂ gas flow rate	0.1 ~ 1.1[sccm]
Input power	30, 45, 60, 75 W
Substrate temp	Room Temperature

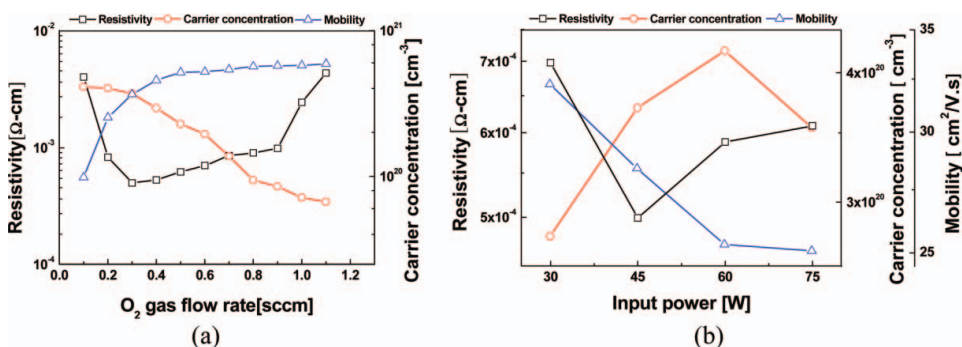


Figure 2. Electrical properties of IZTO, (a) 45W as O_2 gas flow rate, (b) 0.3 sccm as input power

concentration of as-deposited IZTO films decreased with an increasing O_2 gas flow rate, regardless of the input power. When IZTO thin-films were deposited at an input power of 45 W, the carrier concentration of as-deposited IZTO films decreased from 7.30×10^{20} to $6.71 \times 10^{19} \text{ cm}^{-3}$ when the O_2 gas flow rate increased from 0.1 to 1.1 sccm. At a low O_2 gas flow rate, films with a large number of oxygen deficiencies are deposited, which results in high carrier concentrations. As the O_2 gas flow rate increases, the number of oxygen vacancies decreases, which leads to lower carrier concentrations. The resistivity of IZTO thin-films decreased as the O_2 gas flow rate rose from 0.1 to 0.4 sccm, regardless of the input power. When IZTO thin-film was prepared under an O_2 gas flow rate of 0.3 sccm, it exhibited the lowest resistivity of $4.99 \times 10^{-4} \text{ } \Omega\text{-cm}$. On the other hand, when the O_2 gas flow rate exceeded 0.4 sccm, the resistivity of IZTO thin-films increased because the oxygen voids in the thin films were substituted for oxygen atoms and the additional oxygen atoms in the films effectively functioned as carrier traps [12]. Detail comparison information on the other films is shown in Table 2[13].

Figure 3 shows the X-ray diffraction patterns of as-deposited IZTO thin-films at an input power of 45 W and different O_2 gas flow rates. As-deposited films that were deposited

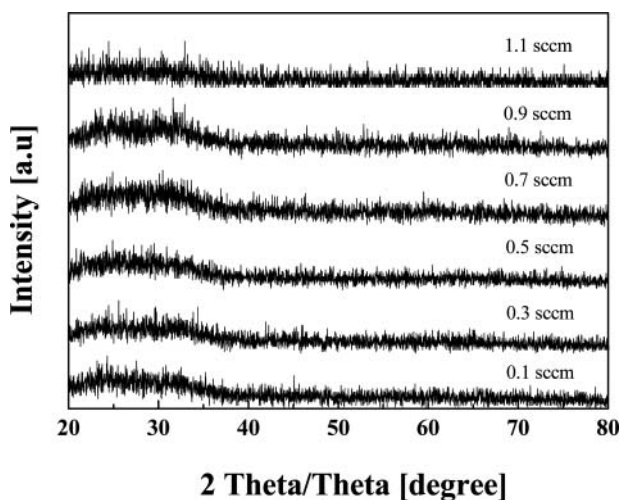


Figure 3. XRD patterns of IZTO at input power 45 W as O_2 gas flow rate

Table 2.

Film Material	A.T. [°C]	P _O [sccm]	T [%]	R [Ω·cm]	μ [cm ² /V·s]	n [cm ⁻³]	Film Growth
ITO	R.T	0.4	65.10	6.05×10 ⁻⁴	21.9	4.39×10 ²⁰	Amorphous
ITO	400°C	0.4	85.59	3.21×10 ⁻⁴	19.3	1.01×10 ²⁰	Polycrystalline
IZTO	R.T	0.3	85.40	4.99×10 ⁻⁴	28.4	3.7×10 ²⁰	Amorphous

A.T. = Annealing temperature
P_O = Partly O₂ gas flow
T = Transmittance at 550 nm
R = Resistivity [Ω·cm]; n = Carrier Concentration [cm⁻³]; μ = Mobility [cm²/V·s]

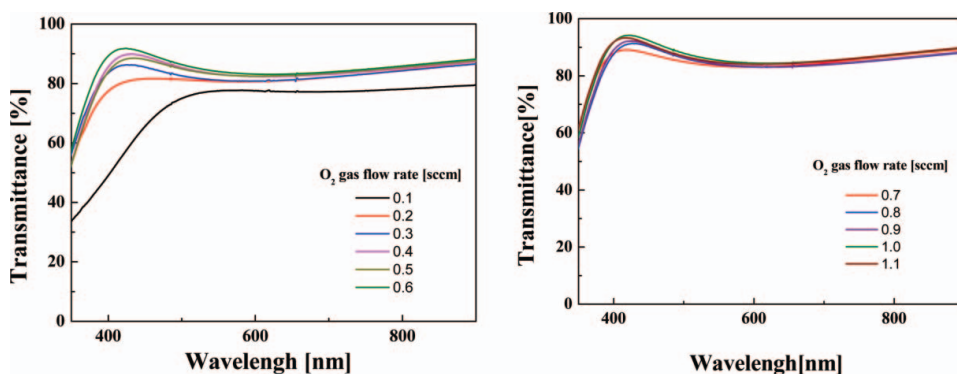


Figure 4. Optical properties of IZTO thin film deposited at 45W as O_2 gas flow rate

on glass did not show any crystalline peaks, regardless of the O_2 gas flow rate. It seemed that the as-deposited IZTO thin-films had insufficient energy for crystallization. This result is attributable to the low deposition temperature and low input power when the IZTO thin-films are prepared [14].

Figure 4 shows the transmittance in the visible range (400–800 nm) of as-deposited IZTO thin-films at an input power of 45 W and various O_2 gas flow rates. IZTO thin-films

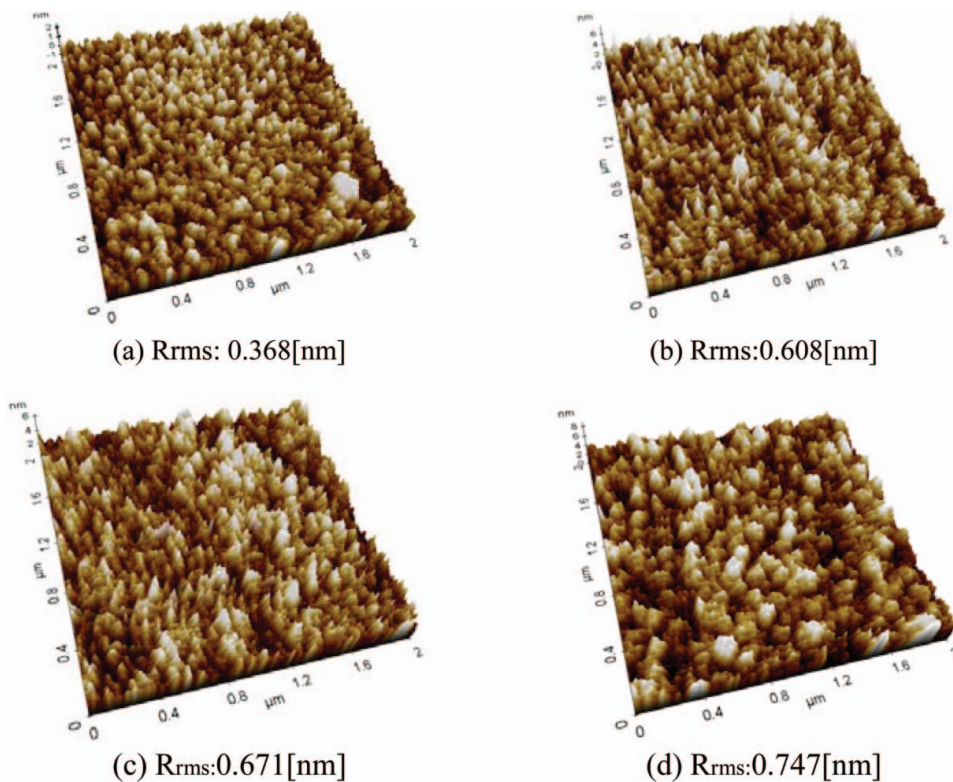


Figure 5. AFM images of IZTO thin films: (a) 30 W, (b) 45 W, (c) 60 W and (d) 75 W

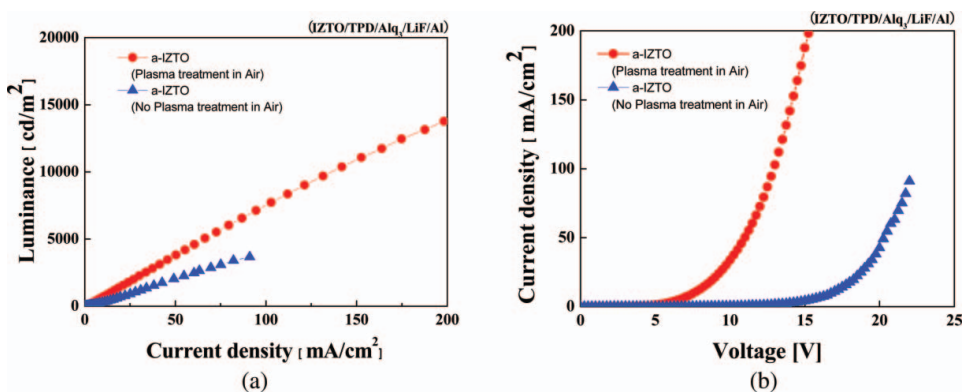


Figure 6. (a) Current density–voltage (J–V) and (b) luminescence–voltage (L–V) of OLED fabricated on a-IZTO anode

showed a transmittance of over 80% in the visible range (400~800 nm) except when the thin film was deposited under an O_2 gas flow rate of 0.1 sccm. The apparent reason was that the transmittance reduced due to an insufficient supply of O_2 gas.

Figure 5 shows AFM images of films that were obtained under a range of the input power. The AFM images were scanned for an area of $2 \times 2 \mu m^2$. Three-dimensional plane views were observed for IZTO films that were deposited under different values of the input power and an O_2 gas flow rate of 0.3 sccm. The Rms surface-roughness values that are calculated from these images are given in Fig. 5. The roughness increases from 0.368 nm to 0.747 nm, as the input power is increased from 30 W to 75 W. It appears that the change of surface roughness in IZTO thin-films is the result of the bombardment of the growth surface by high-energy particles as the input power is increased [15].

Figure 6 shows the J–V–L characteristics of the OLEDs that were fabricated on α -IZTO anode films. As-deposited thin-film was deposited under an input power of 45 W and an O_2 gas flow rate of 0.4 sccm, which yielded the best results. To better ascertain the characteristics of the OLEDs, experiments for two cases, one with plasma surface treatment and the other with no treatment, were separately conducted. As shown in Fig. 5, the turn-on voltage decreased and the current density and luminescence increased when plasma surface treatment was applied to IZTO thin-film than when it was not. The turn-on voltage was 5.5V after plasma surface treatment; in the other case, it was 15.3V. This decreased turn-on voltage was caused by an improvement in the work function and the injection of more holes, which resulted from an increase in the quantum efficiency and the elimination of existing impurities from the surface of IZTO thin-films by plasma treatment [16].

Conclusions

We prepared IZTO thin-films on a glass substrate at room temperature by the FTS method. As-deposited films that were deposited on glass did not show any crystalline peaks, regardless of the O_2 gas flow rate and input power. All IZTO thin-films, except for thin film that was deposited on a glass substrate under an O_2 gas flow rate of 0.1 sccm, had an average transmittance of 80% in the visible range (400–800 nm). The mobility value of IZTO thin-films increased when the O_2 gas flow rate increased from 0.1 to 1.1 sccm; the highest value of the mobility was 51.17 [$cm^2/V \cdot s$] at an input power of 45 W and an O_2 gas flow rate of

1.1 sccm. On the other hand, we observed that the carrier concentration decreased as the O₂ gas flow rate increased; the highest value of the carrier concentration was $7.30 \times 10^{20} \text{ [cm}^{-3}\text{]}$. The resistivity of IZTO thin-films decreased when the O₂ gas flow rate increased from 0.1 to 0.3 sccm; the lowest value of the resistivity was $4.99 \times 10^{-4} \text{ [}\Omega\cdot\text{cm]}$ under an input power of 45 W and an O₂ gas flow rate of 0.3 sccm. When the O₂ gas flow rate exceeded 0.4 sccm, the resistivity of IZTO thin-films increased. The applicability of α -IZTO thin-film as an effective OLED anode is demonstrated. The OLED with a plasma-treated α -IZTO anode shows better J–V–L characteristics than the OLED with a non-treated α -IZTO anode. The turn-on voltage was 5.5V following plasma surface treatment and in the other case, it was 15.3V. The improved performance of OLEDs that were fabricated with a plasma-treated α -IZTO anode could be attributed to the higher work function.

Through FTS, we could obtain IZTO thin-film that could be used for anodes of OLEDs with high transmittance and high mobility at room temperature. In addition, we could prepare IZTO thin-films with low resistivity. It appears that the manufacture of as-deposited IZTO thin-films through the FTS system can be potentially applied for anodes of OLEDs.

Acknowledgment

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