Electrical, optical, and thermal properties of AZO co-sputtered ITO electrode for organic light emitting diodes

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Abstract

In this study, we report on the characteristics of Al-doped ZnO (AZO) co-sputtered indium tin oxide (ITO) films prepared by dual target direct current (DC) magnetron sputtering at room temperature for organic light emitting diodes (OLEDs). The electrical and optical properties of co-sputtered IAZTO electrode were critically dependent on the DC power of AZO. Furthermore, the characteristics of co-sputtered IAZTO electrode were influenced by rapid thermal annealing temperature.

1. Introduction

Indium tin oxide (ITO) film have been widely employed as transparent electrode for hole injection in organic light diodes (OLEDs) and flexible OLEDs due to its high conductivity ($1\text{-}5\times10^3$ S/cm) and optical transmittance (80-85%) in the visible spectra range. Although ITO electrode has been mainly used in OLEDs and flexible OLEDs, it has critical drawbacks as anode materials such as low work function, high process temperature and rough surface [1]. In addition, low transition temperature from amorphous to crystalline of ITO film makes it difficult to apply ITO film as flexible electrode in flexible OLEDs. For this reasons, In-Zn-O (IZO) and In-Zn-Sn-O (IZTO) electrode have gained considerable attention as promising candidates for substituting conventional ITO electrode in OLEDs and flexible OLEDs [2]. In particular, IZTO films have recently been recognized an alternative anode material in OLED and flexible OLEDs due to its high work function (6.1 eV), good conductivity, high transparency, and low deposition temperature [3]. However, the electrical properties of IZO and IZTO electrode were rapidly degraded at high temperature even though they maintain the amorphous structure.

In this work, we have investigated the electrical, optical, structural, and thermal annealing properties of the IAZTO films grown on a glass substrate at constant DC power of ITO target (100W) and variable AZO target power using a dual target sputter system at room temperature. The IAZTO electrode shows comparable resistivity and transparency to those of conventional amorphous ITO electrodes.

2. Experimental

AZO co-sputtered ITO electrodes were deposited on a glass substrate by means of a dual target DC magnetron sputtering system in a pure Ar ambient without the addition of reactive oxygen at a temperature of (~30°C). Both AZO (2 wt % Al_2O_3 doped ZnO) and ITO (10 wt % SnO_2 doped In_2O_3) targets were used and were placed 100 mm from the glass substrate center. For uniform co-sputtering of AZO and ITO targets, we employed a tilted magnetron gun. At constant DC power of the ITO target (100 W), AZO co-sputtered ITO electrodes were grown on a glass substrate as a function of AZO DC power, which ranged from 0 to 100 W for 20 min. In addition, the Ar flow rate and working pressure are maintained constant at 15 sccm and 3 mTorr, respectively. For simplicity, the ITO electrode co-sputtered at AZO DC power of 20 W is referred to as '20 W IAZTO' and so on. Using 20 W IAZTO films grown at optimized conditions, the rapid thermal annealing process was carried out as a function of annealing temperature at base pressure of 5×10^{-6} Torr. The sheet resistance and resistivity of the AZO co-sputtered ITO films were measured by a Hall measurement with van der Pauw geometry at room temperature. The optical transmittance of the AZO co-sputtered ITO films was measured in the wavelength range of 220 to 800 nm by a UV/Visible spectrometer. The structural properties of the AZO co-sputtered ITO films were investigated by x-ray θ-2θ diffraction (XRD) measurements. The extent of AZO

incorporation into the ITO electrode was characterized by Auger electron spectroscopy (AES) using a PHI 670 Auger microscope with an electron beam of 10 keV and 0.0236 μA. The surface morphology of the AZO co-sputtered IZO electrode was analyzed by a field emission scanning electron microscope (FESEM). Finally, the work functions of the AZO co-sputtered ITO films were measured by photoelectron spectroscopy with a UV source (PKI Model AC-2) at atmospheric pressure.

3. Results and discussion

Figure 1(a) shows the sheet resistance, resistivity, and figure of merit (ϕ_{TC}) of the IAZTO electrode as a function of the DC power of the AZO target. A 0 W IAZTO film showed a fairly high resistivity and sheet resistance.

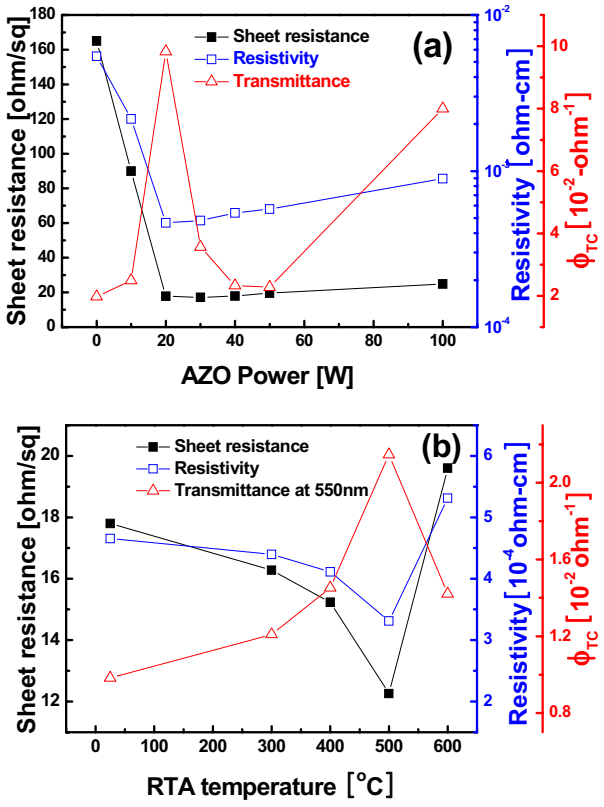

Fig 1. (a) Sheet resistance, resistivity, and figure of merit of IAZTO film as a function of AZO DC power. (b) Sheet resistance, resistivity, and figure of merit of 20 W IAZTO as a function of RTA temperature.

However, co-sputtering of the AZO led to a significant reduction in the resistivity and sheet resistance. The 20 W IAZTO electrode shows the lowest resistivity of 4.6×10^{-4} Ω-cm and sheet resistance of 17.8 Ω /square even though it was prepared at room temperature. Further increase of the AZO DC power above 30 W, however, resulted in a little increase of resistivity and sheet resistance. At AZO DC power of 20 W, the IAZTO films shows the maximum figure of merit values (2.21×10^{-2} Ω^{-1}), which is defined like below [4]

$$\phi_{TC} = \frac{T^{10}}{R_{sh}}$$

where T is the transmittance and R_{sh} is the sheet resistance of the transparent conducting oxide. Figure 1(b) shows the sheet resistance, resistivity, and figure of merit of 20 W IAZTO films as a function of rapid thermal annealing temperature. At the annealing temperature of 500 °C, we obtain much lower sheet resistance (12 Ω/sq) and sheet resistivity (3.3×10^{-4}Ω-cm) than that of as-deposited sample due to the effective activation of dopant element and crystallization of the IAZTO film.

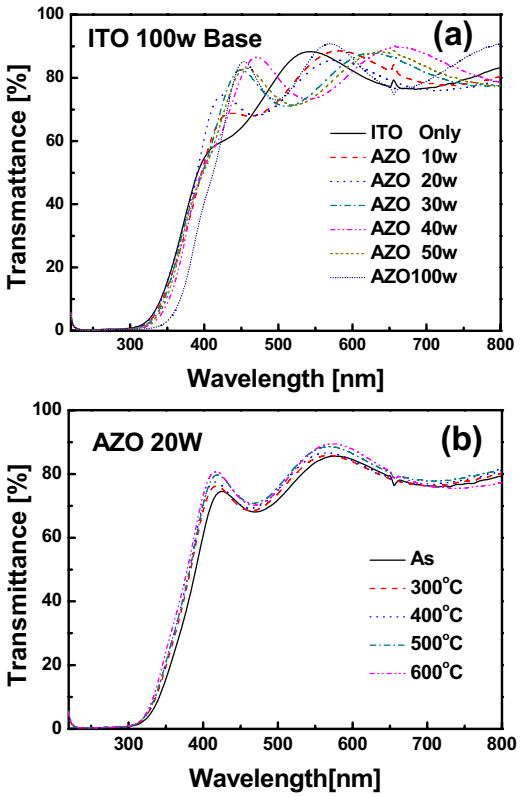

Fig. 2 (a) Optical transmittance of IAZTO films as a function of AZO DC power. (b) Optical transmittance of IAZTO films as a function of RTA temperature.

The transmittance of the IAZTO electrode also depends on the AZO DC power. In contrast to the electrical properties of the IAZTO electrode, the optical transmittance of the IAZTO electrode grown at low AZO DC power, ranging from 10 to 50 W is lower than that of the 0 W IAZTO electrode. However, the 100 W IAZTO film shows comparable optical transmittance to the 0W IAZTO electrode. Figure 2 (b) shows dependence of optical transmittance of the IAZTO film on the temperature of rapid thermal annealing. Compared to as-deposited IAZTO film, the rapid thermal annealed IAZTO film shows improved optical transmittance. The annealed IAZTO film shows better optical transmittance in a visible wavelength regardless of annealing temperature.

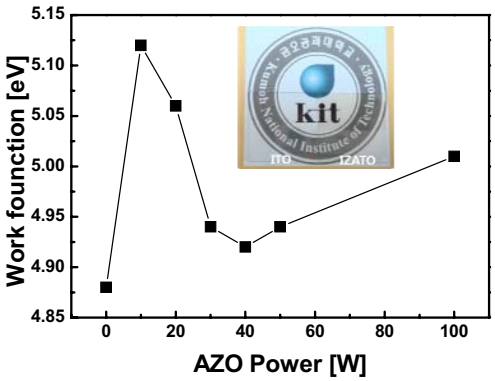

Fig. 3 Work function of IAZTO films as a function of AZO DC power.

Figure 3 shows the work function of the IAZTO electrode as a function of AZO DC power. The work function of the 0 W IAZTO (pure ITO) electrode is 4.78 eV, which is similar to a previously reported value.[5] The 10 and 20 W IAZTO electrodes exhibited significantly increased work functions to 5.12 and 5.06 eV, respectively. However, further increase of AZO power resulted in a decrease of the work function. In order to make high performance OLEDs, it is necessary to employ a TCO anode layer with a high work function greater than 5 eV, since hole injection critically depends on the barrier height formed between the anode and the organic materials. Therefore, the IAZTO anode with a higher work function than that of pure ITO could enhance hole injection into OLEDs. Therefore, the IAZTO anode with a higher work function than that of pure ITO could enhance hole injection into OLEDs. Although the 10 W IAZTO anode exhibits the higher work function, we decided that the 20 W IAZTO anode represents the optimized co-sputtered anode because it has a higher ϕ_{TC} value and lower sheet resistance. The picture in the inset of Fig. 3 shows that the 20W IAZTO anodes have a transparency comparable to the reference ITO anode (0 W IAZTO).

Figure 4 shows x-ray diffraction of the IAZTO electrode as a function of AZO DC power. The 0W IAZTO electrode shows a typical ITO XRD plot with preferred (222) and (400) grain orientation even though it was grown at room temperature. The 10 W IAZTO sample also exhibits crystalline (222) and (400) peaks. It was noteworthy that the XRD plot of the IAZTO electrode changed from polycrystalline to amorphous with co-sputtering of AZO at a DC power above 20 W. It has been reported that the low amorphous/crystalline transition temperature ($T/T_m <$ 0.19 ~ 150 ^oC) of ITO films easily lead to the formation of crystalline ITO as shown in the FESEM image of 0 W IAZTO film. However, ZnO doping into In_2O_3 could maintain a stable amorphous structure below 500^{o}C due to the high amorphous/crystalline transition temperature of ZnO-doped In_2O_3 (~500^{o}C).

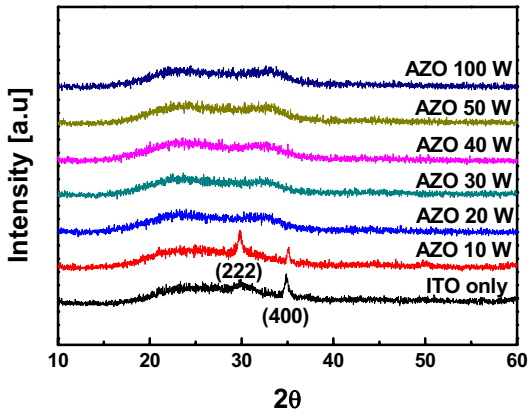

Fig. 4 XRD plots from an IAZTO film as a function of AZO DC power.

Therefore, the amorphous XRD plot of the IAZTO electrodes grown at AZO DC power above 20 W could be attributed to the increase of the amorphous/crystalline transition temperature caused by the introduction of ZnO elements into the ITO matrix.

To investigate the electrical and optical properties of OLEDs fabricated on the IAZTO anode, we prepared conventional OLEDs fabricated on optimized 20 W IAZTO and 0 W IAZTO (ITO) anode layer, respectively. The fabrication process for producing an OLED was described in detail in our previous reports [6]. Figure 5 shows J-V-L

characteristics of OLEDs fabricated on the 20 W IAZTO and ITO electrode grown at room temperature [7]. The current density (J) of an OLED fabricated on the 20 W IAZTO anode is higher than that of an OLED fabricated on an ITO anode at the same voltage after a turn-on of the OLED. The L-V curve of the OLEDs on the 20 W IAZTO anode film also exhibits higher luminance than that of an OLED on the ITO anode as expected from the J-V curve.

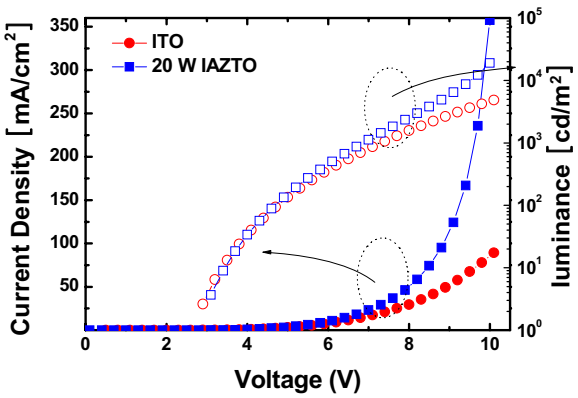

Fig 5. Current density-voltage-luminance (J-V-L) characteristics of an OLED fabricated on 20 W IAZTO and ITO (0W IAZTO) samples.

Due to a lower ohmic loss and the higher work function of the IAZTO layer than the ITO anode grown at room temperature, the OLED on the 20 W IAZTO anode shows higher current density and luminance [3]. The higher work function of the IAZTO layer compared to that of the ITO layer can lead to a lower barrier height between the IAZTO electrode and the organic layer.

4. Summary

Electrical, optical, and structural properties of AZO co-sputtered ITO films on a glass substrate were investigated for application as an anode film in OLEDs. It was found that co-sputtering of AZO with an ITO target resulted in a structural transition from crystalline to amorphous and decreased the resistivity of the IAZTO film. Although an increase of AZO DC power resulted in little decrease of the optical transmittance of IAZTO film, the increase in the work function and the figure of merit value measured in the IAZTO film is beneficial for an anode layer in OLEDs because hole injection from the anode is critically dependent on the work function of the anode. In addition, improved electrical and optical properties of an OLED fabricated on the 20 W IAZTO anode shows the possibility of using IAZTO film prepared by co-sputtering at room temperature as alternative anode materials in OLEDs.

5. References

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