



# Characteristics of Al-doped ZnO films annealed at various temperatures for InGaZnO-based thin-film transistors



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## ABSTRACT

Aluminum-doped ZnO (ZnO:Al, AZO) thin-films were deposited using a pulsed DC unbalanced magnetron sputtering system. The deposited AZO films were annealed in N<sub>2</sub> ambient at various temperatures using a rapid thermal annealing equipment. The influence of the annealing temperature on the structural, electrical, and optical properties of the AZO films was experimentally investigated and the effect of the conductivity of the AZO source/drain (S/D) electrode on the device performance of an oxide-thin film transistor (TFT) was tested. Increasing the annealing temperature resulted in an improvement of the crystallinity of the films. Increasing grain size was found to lead to an increase in the conductivity of the AZO films. The a-IGZO TFTs fabricated with the annealed AZO S/D electrodes showed good performance. Consequently, the performance of the TFT was influenced by the conductivity of the AZO film, which was related to its structural properties.

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## 1. Introduction

Transparent conducting oxide (TCO) films, such as Sn-doped In<sub>2</sub>O<sub>3</sub> (ITO) and III group (B, Al, Ga) doped ZnO, have been widely used for electronic device applications, such as flexible thin film transistors, displays, photovoltaics, and organic light emitting diodes, owing to their high conductivity with a corresponding resistivity of less than  $1 \times 10^{-3} \Omega \cdot \text{cm}$  and high transmittance of over 80% in the visible region [1–4]. The high cost and limited availability of indium, however, limit the cost effectiveness of the conventional indium tin oxide (ITO) electrode as a TCO electrode. Graphene, carbon nanotubes (CNTs) and sheets, and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) have been suggested as promising alternatives to amorphous ITO electrodes for flexible electronic devices [5–7]. However, these materials still have drawbacks in applications as transparent electrodes in cost-efficient flexible electronic devices. Recently, zinc oxide (ZnO)-based films have attracted substantial attention as a potential replacement for the more currently used ITO, due to their low cost, low toxicity, and chemical stability. Aluminum-doped zinc oxide (AZO) films have been extensively explored as a TCO material, owing to their high conductivity, high transparency, low deposition temperature required for producing a crystalline structure, low manufacturing cost, and non-toxicity [8].

In this study, Al-doped ZnO (AZO) thin films were prepared on a glass substrate using a pulsed DC unbalanced magnetron sputtering

(UBMS) system. This sputtering system has various advantages, such as its ability to produce films of superior quality, high growth rate, low deposition temperature, and good uniformity over a large substrate. The effects of the annealing temperature on the crystalline structural, electrical, and optical properties of the AZO films were demonstrated and discussed when they were used as a transparent conductive electrode in an oxide-thin film transistor (oxide-TFT). Also, we fabricated using an AZO film as a source/drain electrode in oxide-TFT devices. Then, we estimated the device performance of the oxide-TFT using AZO electrodes prepared under various conditions, including the drain current–drain voltage ( $I_{DS}$ – $V_{DS}$ ) curve and the drain current–gate voltage ( $I_D$ – $V_G$ ) curves, threshold voltage ( $V_T$ ), on/off ratio, and field effect mobility ( $\mu$ ).

## 2. Experimental details

Al-doped ZnO (AZO) films were deposited on a glass substrate by a pulsed DC unbalanced magnetron sputtering (UBMS) system with an AZO target (99.999%, ZnO:Al<sub>2</sub>O<sub>3</sub> = 98:2 wt.%, LTS Chem.) pressed on a copper saucer with a diameter of 4 in. Prior to the deposition of the AZO film, the process chamber was pumped down to a base pressure of  $7.8 \times 10^{-6}$  Pa using a turbo molecular pump before activating the plasma generation by applying the DC power. The flow rate and working pressure of Ar (99.999%) were fixed at 10 sccm and 0.335 Pa, respectively. Before the deposition of each AZO film, pre-sputtering was performed by an Ar plasma for 5 min to remove the hydrolyzed surface layer of the target. Then, 300-nm-thick AZO films were deposited at room temperature with pulsed DC at a frequency of 20 kHz and duty ratio of 50%. After the AZO films were deposited, they were annealed

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at 300 °C, 400 °C, and 500 °C by rapid thermal annealing (RTA) equipment in nitrogen ambient. The annealing temperature was maintained for 2 min and the rising and falling times were both 30 s at a pressure of  $2 \times 10^{-6}$  Pa. Then, the structural, optical, and electrical properties of the AZO films were investigated with respect to the annealing temperature. To investigate the performance of oxide-TFTs fabricated with the as-deposited and annealed AZO source/drain (S/D) electrodes, heavily doped p-type silicon ( $p^+$  Si) substrates with a low resistivity (conductivity of  $<0.005 \text{ S}\cdot\text{cm}^{-1}$ ) were used as the gate electrodes. The 100-nm-thick  $\text{SiO}_2$  used as the gate dielectric was thermally grown on  $p^+$  Si substrates using a furnace. Then, to investigate the effect of the S/D electrodes, AZO films as the S/D electrodes were deposited by a pulsed DC UBMS system and, in the case of the annealed AZO films, after they were deposited on the  $\text{SiO}_2$  insulator, the devices fabricated as S/D electrodes were annealed by RTA equipment at the respective annealing temperature. Finally, a 50-nm-thick IGZO ( $\text{In}_2\text{O}_3\cdot\text{Ga}_2\text{O}_3\cdot\text{ZnO} = 1:1:1 \text{ mol}\%$ ) film used as an active layer was deposited by using RF magnetron sputtering with a power of 100 W, a working pressure of 0.33 Pa, and mixture gas flow ratio of  $\text{Ar}/\text{O}_2 = 20/0.2 \text{ sccm}$  without substrate heating.

The thickness and surface morphologies of the deposited AZO films were measured using a field emission scanning electron microscope (FESEM: XL-40aFEG, 10 kV) and an atomic force microscope (AFM: Seiko, SPA-400). The crystalline structure of the film was characterized by an X-ray diffractometer (XRD: Bruker, AXS D8 Discover,  $\text{Cu K}\alpha = 0.15405 \text{ nm}$ , 40 kV, 30 mA) in powder diffraction configuration. The XRD patterns were collected in the  $20\text{--}80^\circ 2\theta$  range with a measurement step of  $0.02^\circ$ . The Hall mobility and carrier concentration were examined by a Hall measurement system (ECOPIA, HMS-3000). The spectral transmittances of the films were observed using a UV-spectrophotometer (Hitachi, U 3000) in the visible wavelength range

of 400–800 nm. The existence of Al and Zn atoms and the binding energy of each element in the AZO films were examined by X-ray photoelectron spectroscopy (XPS-PHI5200, 2000 V ion energy, 120 V electron energy, 50 eV pass energy and 0.1 eV energy step size for binding energy) using an Al  $\text{K}\alpha$  radiation source in an ultra-high vacuum. The electrical characterization of the TFTs using AZO S/D electrodes was performed in an ambient atmosphere using an HP4145B semiconductor parameter analyzer.

### 3. Results and discussion

Fig. 1(a)–(d) shows the XPS wide scan and core level spectra of the surface of as-deposited AZO film and AZO film annealed at 500 °C in  $\text{N}_2$  ambient. The XPS wide scan and core level spectra data of the annealed AZO film show  $\text{Al}_{2p}$ ,  $\text{O}_{1s}$ , and  $\text{Zn}_{2p}$  peaks, which are indicative of a bare AZO film. As shown in Table 1, the atomic concentrations of O, Zn, Al, and C obtained in the as-deposited AZO film were 44.9, 34.4, 0.8, and 19.9 at.%, respectively. From Fig. 2(a), a comparison of the  $\text{O}_{1s}$  and  $\text{Zn}_{2p}$  peaks with the  $\text{Al}_{2p}$  peaks showed weak intensity for the latter due to the low doping concentration of  $\text{Al}_2\text{O}_3$  (2 wt.%) in the ZnO matrix. The intensities of the  $\text{Al}_{2p}$ ,  $\text{N}_{1s}$ , and  $\text{C}_{1s}$  spectra for the annealed AZO film were higher than those of the as-deposited AZO film, indicating a larger content of Al and C in the former. The high Al and C content in the film deposited at 500 °C was caused by Zn and O desorption during the RTA process. The presence of a  $\text{C}_{1s}$  peak in the spectra can be attributed to contamination, which resulted from the samples being exposed to the ambient atmosphere [9]. However, the AZO film annealed at a temperature of 500 °C exhibited atomic concentrations of 40.5% O, 27.5% Zn, 1.0% Al, and 30.3% C. Compared with the results of the as-deposited AZO film, the atomic concentrations of O and Zn in the annealed AZO film were decreased, and the concentrations of Al and N atoms were

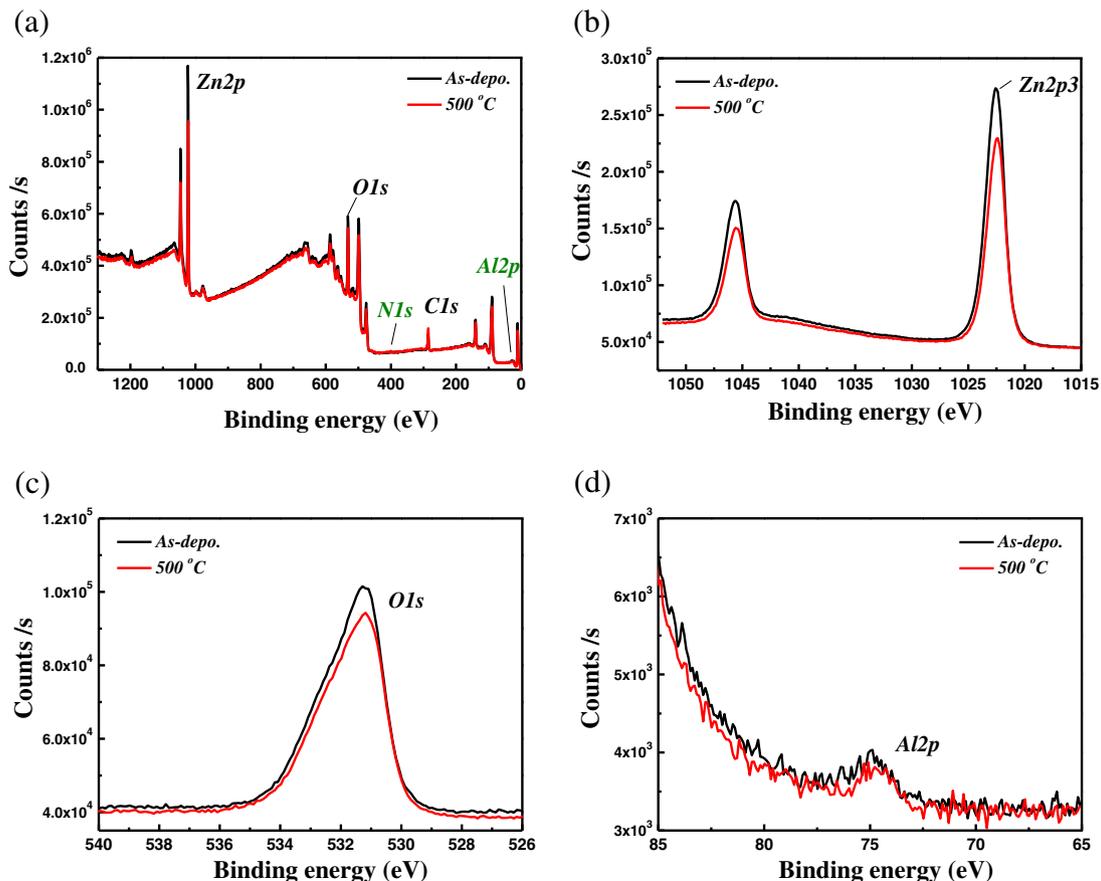


Fig. 1. (a) XPS wide scan, and (b–d) core level spectra of  $\text{Al}_{2p}$ ,  $\text{O}_{1s}$ , and  $\text{Zn}_{2p}$  of AZO film annealed at 500 °C temperature.

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