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The low temperature synthesis of Al doped ZnO films on glass and polymer using pulsed co-magnetron sputtering: H2 effect

Yun M. Chung *, Chang S. Moon, Woo S. Jung, Jeon G. Han

Center for Advanced Plasma Surface Technology, Sungkyunkwan University, 300 Chunchun-dong, Jangan-gu, Suwon 400-746 Korea

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Abstract

It has been reported that a small amount of hydrogen in argon plasma induces an increase in the crystallite size of the as-deposited films. In addition, control of the hydrogen partial pressure is expected to improve the carrier mobility by increasing the crystallinity of the film (larger crystal size and lower grain boundary effects). Al doped ZnO (AZO) films were deposited by co-CFUBM (closed field unbalanced magnetron) sputtering. The ultimate aim was to deposit transparent films on a polymer substrate with a low electrical resistivity. Therefore, the structural, optical and electrical properties of AZO films were investigated as a function of the hydrogen partial pressure. A minimum resistivity and maximum transparency of 8×10^{-4} Ω cm and 88.1% were obtained, respectively. A critical P_{H2} was expected to improve the carrier mobility by increasing the crystallinity of the film. However, above this value, conductivity reduced due to the formations of oxides such as ZnO and Al_2O_3 in the AZO films.

Keywords: Al doped ZnO; Hydrogen dilution process; Polymer; Flexible device

1. Introduction

The development of thin coatings with a high transmittance and low resistivity is an important issue in photovoltaic and optoelectronic applications such as flat panel displays and solar cells. In addition, the low temperature synthesis of transparent conductive oxide (TCO) film on polymer had attracted a great deal of interest because flexible electronic device applications are becoming a new market in the electronic industry. However, a high temperature process or substrate bias control is needed to obtain good electrical and optical properties due to the microstructural characteristics of TCO [1–7]. This has limited the application of transparent TCO electrodes to flexible displays. The main problem for depositing films on a polymer is that the substrate temperature needs to be relatively low. However, under this condition, the sputtered ions from the targets cannot obtain sufficient heat energy from the substrate to adjust the bond direction and length in order to obtain the optimum bonding to the adjacent atoms. This leads to bad adherence and as well as a low mobility and a high resistivity as a result of the strong grain boundary scattering of the charge carriers [8].

In previous work, a co-CFUBM sputtering process was proposed for the low temperature synthesis process of TCO, and Al doped ZnO (AZO) films were deposited as a substitute material for indium tin oxide (ITO) on glass and polymer substrates at low temperature <50 °C with a controlled working pressure, P_{Ar} [9], oxygen partial pressure, P_{O2} [10] and Al content [11]. A minimum resistivity and maximum transparency of 1.2×10^{-3} Ω cm and 84.2% were obtained, respectively. However, these properties are unacceptable for industrial applications. Therefore, this study introduced a hydrogen dilution step in the co-CFUBM sputtering process for the synthesis of highly conducting, transparent and more stable AZO films. The structural, electrical and optical properties of the AZO films were investigated as a function of the hydrogen partial pressure, P_{H2}.

2. Experimental details

The deposition chamber was equipped with two home-made unbalanced magnetron sputtering sources (Φ 2 in.) equipped a ZnO target and an Al target for the low temperature process, which were designed to negate the

* Corresponding author. Tel.: +82 31 299 6644; fax: +82 31 290 5669.
E-mail address: ymchung@skku.edu (Y.M. Chung).

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Table 1
Conditions for AZO co-deposition

Deposition parameters	Conditions
Base pressure	0.004 Pa
Ar partial pressure	2.7 Pa
Hydrogen partial pressure	0~4.0 Pa
Input power	Unipolar pulsed DC mode
ZnO cathode	Frequency: 30 kHz
	Duty cycle: − duty 50% and + duty 45%
	Power density: 30 W/cm^2
Al cathode	DC mode
	Power density: 3.5 W/cm^2
Total film thickness	200 nm
Distance from target to substrate	8 cm
Jig rotation speed	20 rpm
Ambient temperature	50 °C

unstable transition region between the metallic and compound deposition mode and to allow for the easy control of the Al content [9,11]. The Al doped ZnO films with a thickness of 200 nm were deposited on glass (Corning 1737) and polymer (PES, polyethylene sulfate) with a 20 rpm rotation at an ambient temperature of 50 °C. A detailed description of the experimental parameters is given in Table 1. From previous work, the ZnO and Al cathode power density were fixed to 3.5 and 30 W/cm^2, respectively. The microstructure of the AZO films was investigated by X-ray diffraction (XRD) and transmittance electron microscopy (TEM). The bonding structure was analyzed by X-ray photoelectron spectroscopy (XPS). The electrical and optical properties are measured using a four point probe and UV–visible spectroscopy, respectively.

3. Results and discussion

Fig. 1 shows the spatial dependence of the film resistivity on the hydrogen partial pressure. At a P_{H2} of 0.13 Pa, a minimum resistivity of 8×10^{-4} Ω cm and 1×10^{-3} Ω cm were obtained for the AZO:H films on glass and polymer, respectively. In previous work [10], better conductivity was obtained at the substrate position of 3 cm outside from the target center due to the incident angle of the adatoms that enhanced the adatom mobility. The substrate position corresponding to the target center (heavy bombardment area

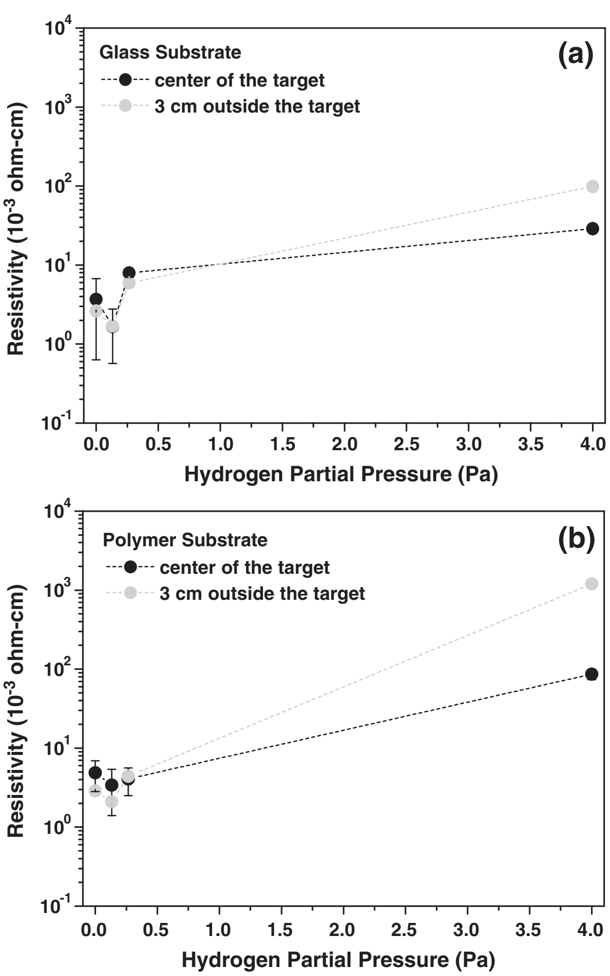

Fig. 1. Electrical resistivity of the films on glass (a) and polymer (b) plotted as a function of the hydrogen partial pressure.

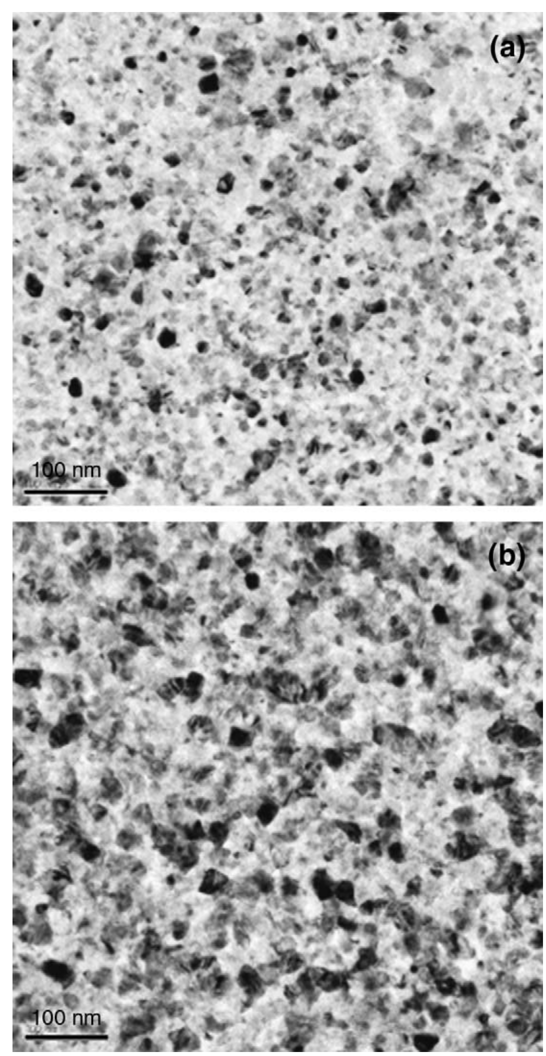

Fig. 2. Plane-view transmission electron micrographs of AZO (a) and AZO:H (b) films with a film thickness 150 nm.