

# Growth of Aluminum Doped Zinc Oxide Films on Polymer Substrates for Flexible Display Applications

Jae-Hyeong Lee and Jong-In Lee

**Abstract**—Highly conductive and transparent aluminum doped ZnO thin films (AZO) films have been prepared by r.f. magnetron sputtering processes on polycarbonate (PC) and onto glass as reference. In addition, the electrical, optical properties of the films prepared at various sputtering powers were investigated. The XRD measurements revealed that all of the obtained films were polycrystalline with the hexagonal structure and had a preferred orientation with the c-axis perpendicular to the substrate. The ZnO:Al films were increasingly dark gray colored as the sputter power increased, resulting in the loss of transmittance. High quality films with the resistivity as low as  $9.7 \times 10^{-4} \Omega\text{-cm}$  and transmittance over 90% have been obtained by suitably controlling the r.f. power.

**Index Terms**—Al doped ZnO, Thin film, Polymer substrate, Flexible display

## I. INTRODUCTION

Zinc oxide, one of the most important binary II–VI compounds, is a direct semiconductor of wurtzite structure. Its minimum energy gap is 3.2 eV at room temperature and 3.44 eV at 4K [1]. Compared to Sn-doped In<sub>2</sub>O<sub>3</sub> (ITO), it has a number of advantages, such as non-toxicity, low cost, material abundance, and high stability against hydrogen plasma and heat cycling [2].

Transparent conducting films deposited on polymer substrates have many merits compared with those deposited on glass substrate: they have a light weight, a small volume and the obtained devices can be folded, easy to carry [3]. It is well known that transport and optical properties of sputtered AZO films are highly sensitive to processing parameters, particularly, sputtering power, deposition pressure, and oxygen concentration in the working gas. In this work, AZO films on PET substrates have been prepared by magnetron sputtering method using powdery target with different deposition conditions. The structural, optical and electrical properties of the obtained films depending on sputtering power have been investigated.

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## II. EXPERIMENTAL

The AZO films were deposited on Corning 7059 glass and polycarbonate (PC) substrates in a conventional r.f. magnetron sputtering system. A sintered oxide ceramic disk of ZnO (diameter 3 inch) mixed with 2 wt.% Al<sub>2</sub>O<sub>3</sub> was used as a target. The base pressure in the sputter chamber was below  $6.6 \times 10^{-4}$  Pa. The separation between target and substrate was about 6 cm. After the chamber was evacuated to a base pressure below  $6.6 \times 10^{-4}$  Pa, pre-sputtering of 20 min was carried out at an argon gas pressure of 1.0 Pa in order to clean the target surface. The sputtering power was varied from 50 to 200W, and the argon gas pressure was controlled from 0.3 to 2.7 Pa. All the films were deposited at room temperature. In order to reveal the genuine effect of r.f. power excluding the film thickness effect, we have fixed the film thickness to about 200 nm.

## III. RESULTS AND DISCUSSION

AZO films prepared in this work were physically stable and had good adherence to the polymeric substrate. Fig. 1 shows the relation between the sputtering power and deposition rate of AZO films on glass substrate. A linear increase on growth rate was observed as sputtering power increase. This increase indicates the number of atoms sputtered from the target is proportional to the r.f. power. For higher sputtering power, the sputtered species get a higher energy that contributes to the film growth. These high energy particles have high surface mobility and therefore a higher growing process at the surface takes place.

Fig. 2 shows the XRD patterns for AZO films deposited on glass and PC substrate at several r.f. powers (ranging from 50 to 125 W). AZO films on glass have a strong diffraction peak at  $2\theta \sim 34.4^\circ$  and very weak peak at  $2\theta \sim 72.5^\circ$ , regardless of sputter pressure and substrate types. These peaks are associated with the (0 0 2) and (0 0 4) plane of hexagonal phase ZnO. The presence of a strong diffraction peak of (0 0 2) indicates that films have a (0 0 2) preferred orientation. Neither metallic zinc or aluminum characteristic peaks nor aluminum oxide peak was observed from the XRD patterns, which implies that aluminum atoms replace zinc in the hexagonal lattice or aluminum segregate to the noncrystalline region in grain boundary. With increase-

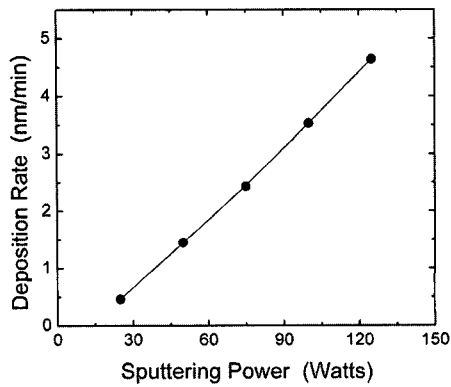


Fig. 1 Influence of sputtering power on the deposition rate of AZO films on glass substrate.

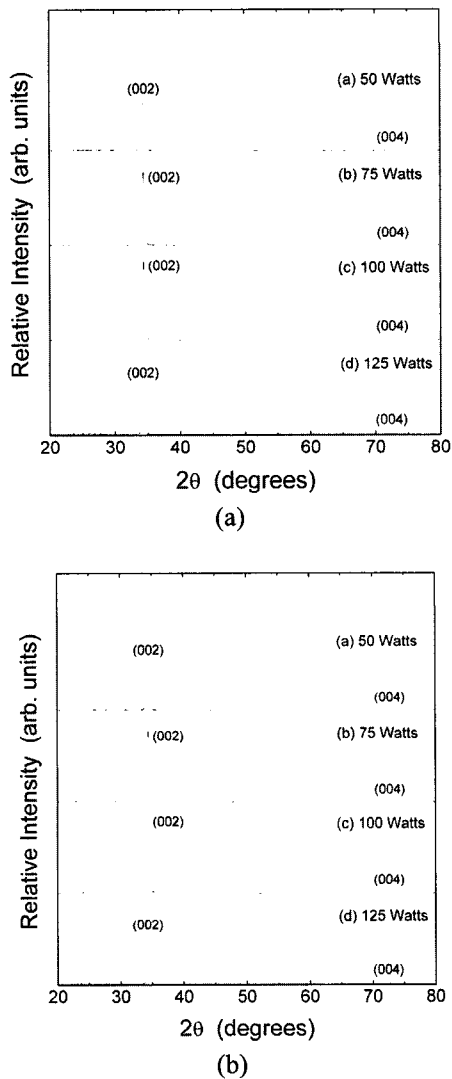


Fig. 2 X-ray diffraction patterns for AZO films deposited at different sputtering powers: (a) glass substrate; (b) PC substrate.

ing the sputtering power the intensity of the (0 0 2) peak increases. This means that the crystallinity of the resulting films improves when the r.f. power is higher. It is noteworthy that the intensity of (002) diffraction peak decreases when the sputtering power exceeds 100 W.

High r.f. power induces faster reaction rate and it damages the surface, resulting to a poor crystalline quality. Accordingly, the sputtering power must be sufficiently low to have an efficient nucleation and growth. For PC substrate, the XRD spectra are very similar to those of glass substrate. This means that the films deposited on PC have the same structure, and both of them are polycrystalline with a hexagonal structure and a preferred orientation with the c-axis perpendicular to the substrates. By comparing the results with the same film deposited onto glass substrates, however, the intensity of the diffraction peaks for PC substrate is not so intense, which might be due to the surface roughness exhibited by the polymer substrate.

The SEM photographs of AZO thin films with various sputtering powers are shown in Fig. 3. The morphology of AZO grains is found to be continuous and dense. The surface grain size tends to be bigger when r.f. power increases, regardless of substrate types. High r.f. power allows that the deposited particles may grow with bigger grains, improving thus the surface state density. Besides, the particles have higher kinetic energy, being obtained more compact films. However, Kim *et al.* [4] reported that the grain size of the ZnO film is not much dependent on the r.f. power in the range of 80–250 W, even though there is a slight increase in the grain size at higher r.f. powers. They concluded that the temperature elevation at higher r.f. power helps to obtain the slightly larger grain structure. The crystallite sizes of the films deposited on PC substrates are larger than that on glass under the same deposition conditions, as seen in Fig. 3 (b) and (d).

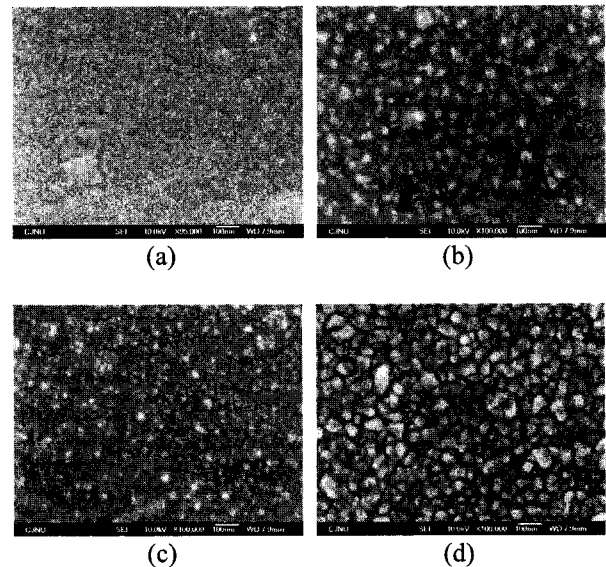


Fig. 3 SEM micrographs of AZO thin film deposited at various sputtering powers: (a) glass substrate, 50W; (b) glass substrate, 125W; (c) PC substrate, 50 W; (d) PC substrate, 125 W.

Fig. 4 illustrates the resistivity, carrier concentration, and Hall mobility of AZO deposited on glass and PC substrate as a function of sputtering power. It can be seen that as the sputtering power increases from 25 to 125 W,

the resistivity of the films on glass decreased significantly from  $5.8 \times 10^{-2}$  to  $9.7 \times 10^{-4} \Omega\text{-cm}$ . This behavior could be explained by the effect of the sputtering power on the mobility and carrier concentration (Fig. 4(b)). When the sputtering power increases, the carrier concentration increases slightly while the mobility increases significantly. These variations originate from improved crystallinity and enhanced substitutional doping as the sputtering power increases. The former effect increases the crystallite size and, subsequently, reduces charge carriers scattering due to grain boundaries, thus increasing the mobility, while the latter effect increases the carrier concentration. Both of them reduce the resistivity of the films. The maximum value of the Hall mobility was obtained for the r.f. power of 100 W and then a slight decrease was observed. This could be attributed to the increase in the scattering from ionized impurities. The low value of the mobility obtained at 25 W is related to the fact that the films were deposited at room temperature. At low temperature, the sputtered ions from the target cannot obtain enough heat energy to adjust the bond direction and length in order to obtain an optimum bonding to the adjacent atoms (including substrate atoms and film atoms). This leads to

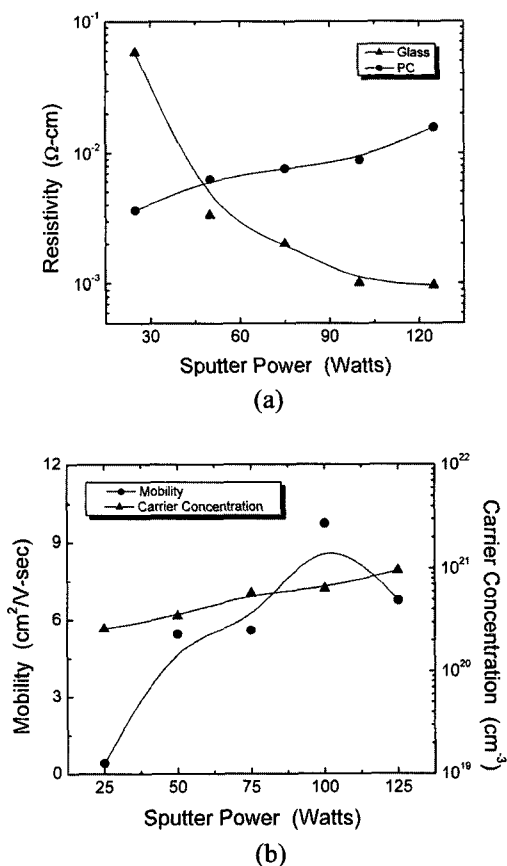


Fig. 4 Dependence of electrical properties of ZnO:Al films on sputtering power: (a) resistivity; (b) carrier concentration and Hall mobility. The substrate type is glass.

difficulties, not only due to a nucleation on the substrate

for the sputtered ions and ion clusters, but also to a growth of large grains from the nucleated molecules. The former effect leads to a bad adherence to the substrate for obtained films and the latter causes a low mobility and a high resistivity due to a strong contribution of grain boundaries in charge carriers scattering. For PC substrates, meanwhile, the resistivity increases with the sputtering power, as seen in Fig. 4. At high r.f. power, enhanced high energetic ion bombardment leads to damage of organic substrate, and the quality of ZnO thin film will become poorer. Accordingly, the sputtering power must be sufficiently low to have an efficient nucleation and growth for organic substrate.

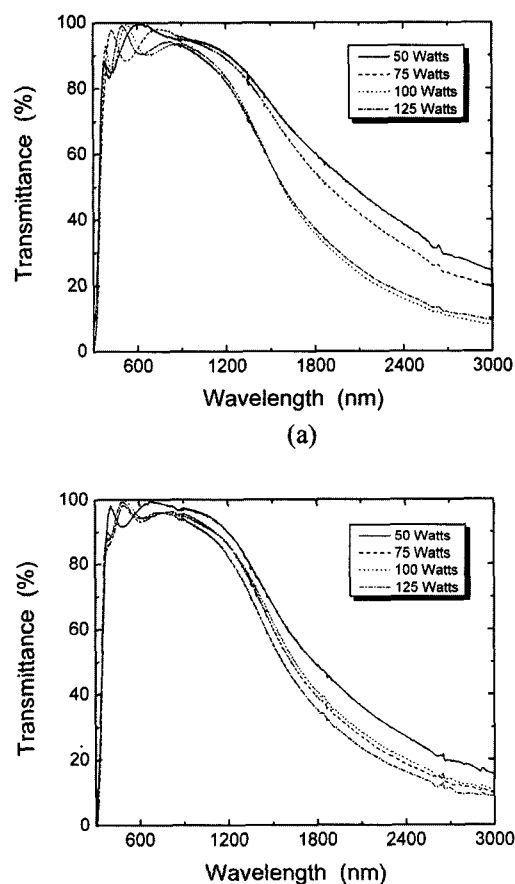


Fig. 5 Optical transmittance of AZO films deposited at various sputtering powers: (a) glass substrate; (b) PC substrate.

Fig. 5 shows the optical transmittance of AZO films deposited at various sputter powers. The optical transmittance spectra of AZO films represent a strong dependence on the sputter power. It is seen that the average transmittance of these samples in the range 400–800 nm was over 90%, regardless of sputtering voltage and substrate types. Fortunato *et al.* [5] reported that AZO films deposited on PET substrate showed the average transmittance of 85% in the visible part of the spectrum. As the r.f. power increases, the average transmittance of the films reduces slightly. It would be noted that the film deposited at 125 W becomes nearly

opaque to near-infrared region. However, the dependence of substrate types on transmittance is not as sensitive as the resistivity's.

#### IV. CONCLUSIONS

AZO films were deposited on glass and PC substrate by r.f. magnetron sputtering at room temperature. The obtained films were polycrystalline with the hexagonal structure and had a preferred orientation with the c-axis perpendicular to the substrates, regardless substrate types. The improvement in the electrical properties is obtained by the increase of the r.f. power essentially due to a substantial increase in the carrier concentration and Hall mobility. This behavior could be related to the increase in the sputtering rate and also to the enhancement in the crystallinity of the films. The optical transmittance of the films was over 90% in the visible region, regardless of r.f. power and substrate types. The transmittance in the near-infrared region reduced with the r.f. power. Although the quality of the AZO films deposited on organic substrates is somewhat worse than that for glass substrate, AZO films on PC might be suitable for solar cells.

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Was born in Seoul, South Korea, on July 5, 1970. He received the B.S. and M.S. degree in electrical engineering from the Sungkyunkwan University, Suwon, South Korea in 1991 and 1994, and the Ph.D. degree in II-VI compound solar cells from the same University in 1998. He spent four years as a senior researcher at Science and Technology Institute of Sungkyunkwan University. From 2002 to 2003, he was a research professor at School of Information and Telecommunication, Sungkyunkwan University. He joined the Kunsan National University in 2003, where he is currently Professor. He has authored and coauthored more than 60 papers at domestic and

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Was received the B.S. degree from Ulsan University in 1979, the M.S. and Ph.D. degree from Chonbuk University in 1983 and 1987, respectively, all electronic engineering. In 1988, he joined Kunsan National University, Chonbuk, Korea, where he is currently a Professor in School of Electronic and Information Engineering. From 2003 to 2004, he held a research position with the Department of Electronic Engineering, Sydney Australia. His interests include the analysis and synthesis of network, analog and digital filter circuit design and semiconductor process and circuit design.