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Microstructure, opotoelectrical and pre-strain dependent electrical properties of AZO films on flexible glass substrates for flexible electronics

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ABSTRACT

Al-doped zinc oxide (AZO) thin films were deposited on ultra-thin flexible glass substrate using a direct current magnetron sputter. Microstructure, electrical and optical properties of the AZO films were investigated as a function of film thickness. The XRD pattern shows that all deposited AZO films have a highly preferred c-axis orientation along (0002) direction. When the film thickness is over 100 nm, all the AZO films have similar grain size about 30 nm and good uniformity. The sheet resistance of the AZO films decreased with the increase of film thickness, whereas the 230.1 nm thick AZO film has the minimum resistivity of 1.65×10^{-3} Ω cm. The average transmittance of all AZO films in the visible range was about 80%, while the optical band gap of the films was in the range of 3.483–3.526 eV. The sheet resistance of AZO films deposited on flexible glass using strained growth is different with that of AZO films using normal growth method. Moreover, the sheet resistances of AZO film with different pre-strain are very different, which may be due to their different microstructures of AZO films.

1. Introduction

Transparent conductive oxide (TCO) thin films are widely used in many areas such as sensors [1], organic light-emitting diodes (OLEDs) [2], electrodes [3], solar cells [4], etc. Indium tin oxide (ITO), fluorine doped tin oxide (FTO) and aluminum-doped zinc oxide (AZO) are well-known TCO materials because of their excellent electrical and optical properties [5–7]. At present, much attention has been paid to investigate the AZO thin films due to their low cost, non-toxicity, good thermal stability and resource abundance [8–10]. The AZO thin films have been deposited on various polymers (including polythylene naphthalate (PEN), polythylene terephthalate (PET) and polycarbonate (PC)) [11–13] However, these flexible substrates are easily degraded by substrate heating during film deposition [2]. Glass substrates are transparent and can endure high temperatures [14], but they are normally rigid thus unsuitable for flexible electronic applications. Therefore, polymer and glass substrates limit the wide applications of the AZO thin films into flexible, transparent and high-temperature conditions.

Recently Corning Incorporated has developed an ultra-thin flexible glass, named Corning® Willow® Glass [15]. Willow Glass is thin enough to be flexible but still can retain its superior features with the same thickness as a sheet of printing paper. Willow Glass has a good heat resistance and its processing condition can be up to 500 °C which cannot be realized using polymer films. Although many applications based on the Willow Glass have been investigated including displays [16], thin film transistors [17], surface acoustic wave strain sensors [18] and photovoltaics [19], there are few references on the investigation of the optical and electrical properties of AZO thin films on Willow Glass substrates. To the best of our knowledge, only Peng et al. investigated the effect of the strained growth and normal growth methods on the electrical and optical degradation of the AZO thin films on Willow Glass under cyclic bending conditions [20]. However, their work did not give the influence of sputtering condition on the opotoelectrical properties of AZO films.

In this current work, we present a systematic study of the effect of film thickness on the microstructure, surface morphology and electrical and optical properties of AZO thin films grown on flexible Willow Glass substrates. At last, the pre-strain dependent electrical properties of AZO films on curved glass substrates were investigated and discussed.

2. Experimental details

AZO thin films were deposited on 200 μm thick Willow Glass substrates using a direct current (DC) sputtering method with a AZO target (ZnO 98 wt%: Al_2O_3 2 wt%, 60 mm diameter and 4.5 mm thickness). In the experiments, the base pressure was 6.0×10^{-4} Pa, the DC power was 100 W and the working pressure was 1.0 Pa. The Ar gas flow rate was kept at 30 sccm throughout the deposition condition and the substrate was under room temperature. The sputtering time was varied from 10 to 60 min in steps of 10 min. Correspondingly, the film thicknesses are 47.0 nm, 128.8 nm, 155.7 nm, 230.1 nm, 282.1 nm and 342.6 nm, respectively. Before deposition, a pre-sputtering of 15 min

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was performed to clean the target surface. These six samples are represented by the symbols of T10, T20, T30, T40, T50 and T60 in the following, respectively. After film deposition, the microstructures of the AZO films were characterized by X-ray diffraction (XRD, MAC, M18XHF) with a Cu-Ka radiation source ($\lambda = 1.5418$ Å). The thicknesses of the AZO films were measured using a stylus profiler (BRUKER, Dektak-XT). Surface morphologies of the AZO films were analyzed using scanning electron microscopy (SEM, Hitachi, S4800). The sheet resistance of the AZO thin film was measured using a four-point probe method (PROBES TECH, RTS-9). The transmittance spectra and the absorption spectra of the AZO films were investigated using ultraviolet–visible spectroscopy (Shimadzu, UV-2550).

3. Results and discussion

3.1. Microstructure analysis

Fig.1(a) shows the XRD patterns of the AZO films with different thicknesses. The intensity of the T10 sample is very weak comparing with other samples, which indicates that 10 min is not enough for the formation of the crystalline AZO film. Then the intensity of the diffraction peak becomes stronger and stronger with increasing the film thickness. All the diffraction peaks are located near 34.4°, which shows that all the sputtered AZO films have wurtzite phases with a highly preferred (0002) orientation. To obtain the detailed structural information, the mean grain sizes of the AZO thin films were calculated by the Debye–Scherrer equation [21] as given in the following:

$$D = \frac{0.89\lambda}{\beta \cdot Cos\theta} \tag{1}$$

where D is the grain size, λ is the X-ray wavelength, β is the full width at half maximum (FWHM) of X-ray peak, θ is Bragg diffraction angle. The calculated FWHM and the grain size versus film thickness are given in Fig.1(b). The FWHM value of T10 sample is the biggest and so its grain size is smallest since the grain size is inversely proportional to the FWHM. As the film thickness increases, the FWHM value decreases and the grain size increases. These last four samples have similar FWHM, which results to their similar grain size.

Surface morphology of TCO thin films is critical to affect their opotoelectrical applications. Fig. 2 shows the SEM images of AZO thin films with different thicknesses. It shows that an obvious increase of grain size is observed and the film's uniformity is improved when the film thickness increases from 47 to 128.8 nm. As the thickness increases from 128.8 to 342.6 nm, the grain size in Fig. 2(b), (c), (d), (e) and (f) has no significant changes. These results are in good agreement with the XRD measurements.

3.2. Electrical properties

The sheet resistance of AZO thin films was measured by using a four-point probe method and the results are shown in Fig. 3. It can be seen from Fig.3 that the sheet resistance of the films drops greatly from 673.3 Ω/sq to 51.7 Ω/sq with increasing AZO film thickness from 47.0 nm to 342.6 nm. This trend of the sheet resistance's change is in agreement with the work of Zhou et al. [22]. In addition, the resistivity of the film can be calculated by using the following formula:

$$\rho = R_S \times d \tag{2}$$

where ρ is the resistivity, R_S is the sheet resistance and d is the thickness of the films. The effect of film thickness on the electrical resistivity of AZO films is also shown in Fig.3. It shows that the resistivity decreases from 3.16×10^{-3} Ω cm to the minimum of 1.65×10^{-3} Ω cm and then increases a little with the increase of film thickness. As it is mentioned earlier that higher film thickness leads to the higher crystallinity of the AZO films as seen from XRD results. Enhancing the crystallinity of AZO films can increase the grain size, decrease the grain boundary scattering and then increase the carrier's lifetime, which results in lower resistivity of AZO films [23]. However, when the film thickness increases further, the too high carrier concentration in AZO film results in lower electron mobility caused by scattering centers in grain boundaries [24]. As a result, the resistivity of AZO film decreased a little when the film thickness still increases. So the AZO film with 230.1 nm thickness has the smallest resistivity of 1.65×10^{-3} Ω cm in our experiments.

3.3. Optical properties

Fig. 4(a) shows the transmittance spectra of the AZO films in the range of 200–900 nm. It is clear that all AZO films exhibit an apparent absorption edge at about 330 nm and the absorption edge demonstrates a red-shift with the increase of film thickness. Meanwhile, all the AZO films demonstrate a high transmittance in the visible range and the average transmittance values of the AZO films with different thicknesses in the visible range are 77.3%, 86.8%, 80.8%, 83.5%, 83.9% and 85.5%, respectively. The surface scattering of thin film maybe plays an important role in reducing the light transmittance. In addition, the wave fluctuation is observed in almost every transmittance spectrum. Moreover, the number of the peaks and the troughs of the curves gradually rises with the increase of film thickness. This similar phenomenon is also found in other AZO films [23] and Ta_2O_5 films [25] with different thicknesses, which is mainly due to the interference effect of the transmitted light with different wavelength.

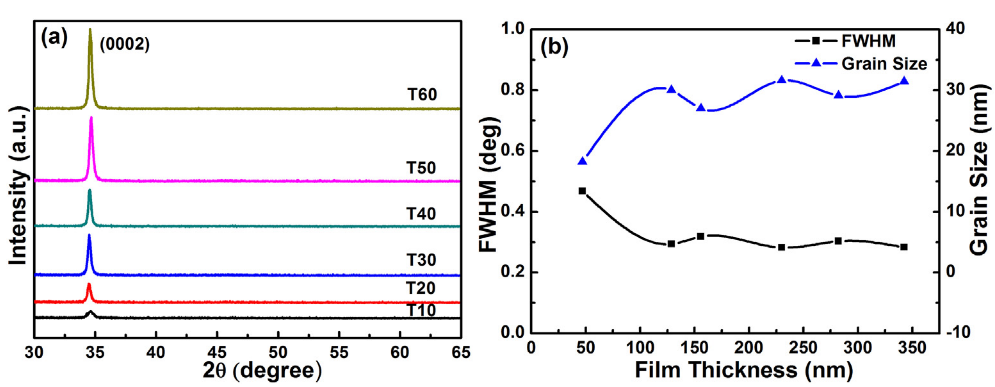

Fig. 1. XRD pattern (a) and corresponding FWHM and grain size (b) of AZO thin films with different thicknesses.

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