Physical properties of ZnO:AI films prepared at low temperature in high power on flexible substrate by RF sputtering

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1. Abstract

Transparent and conductive aluminium-doped zinc oxide (AZO) thin films were deposited on a polyethylene terephthalate (PET) substrate at a low temperature by radio-frequency sputtering using an AZO target composed of ZnO:Al2O3 (2 wt.%). We report a method to deposit in high power radio frequency of ZnO:Al (AZO) films on PET substrate without any crack and low resistivity. The sputtering was carried out in an Ar gas (99.99%) atmosphere with varying RF power ranging from 100 to 300 W. The effects of RF power on morphology, structural, resistivity and optical properties in low and high RF power were investigated. The structural properties of as-deposited films were analysed by X-ray diffraction managements (XRD) confirmed that films have hexagonal wurtzits structure and a strong preferred c-axis orientation (002). Resistivity as low as $1.1*10^{-3}$ Ω cm was achieved for the film deposited at 250 W and 0.2 Pa. High optical transmittance (95%) was exhibited when films were deposited at RF powers below 150W. Band gap energies ranged from 3.36 to 3.39 eV.

2. Introduction

Transparent conducting films on flexible substrates have been investigated extensively because their interesting electrical and optical properties make them suitable for many applications [1–3]. They are used in flexible electro-optical devices, plastic liquid crystal display devices, heat reflecting mirrors and heterojunction solar cells. Al-doped ZnO (AZO) films have attractive much attention because of their comparable high optical transmittance and low electrical resistivity with respect to other TCOs widely used such as Tin-doped Indium Oxide (ITO) films. Although ITO films use as a most successful TCO because of its optical and electronic properties but it need to replaced due to high production cost, non-compatibility to all flexible substrates, its toxicity and brittle behaviour. For this reason AZO can be considered as a very promising candidate because of its low cost, high electrical conductivity, piezoelectricity, easy fabrication, non-toxicity and many application advantages[4].

There are several deposition method to grow AZO films such as chemical vapor deposition, spray pyrolysis[5,6], pulsed laser deposition [7], sputtering method[8,9] and sol gel [10]. AZO films showed low values of resistivity at low temperature deposition because of radio-frequency sputtering is considered as a favourable deposition technique. This techniques effective method of depositing TCO films due to large area uniformity, low temperature deposition and the films show good adhesion on substrates.

In this work, we report on the successful deposition of AZO layer on PET polyester foil of 125 µm in high power RF without any crack. attempt was to improve the electrical properties of AZO thin films deposited on flexible substrate. The dependence of the electrical, structure and optical properties on AZO films on the power was investigated. The choice of the substrate, polyethylene terephthalate (PET), was based on the fact that this is a unique polyester that maintain sits physical, mechanical, electrical and chemical properties for working temperatures up to150 °C.

3. Experimental procedure

AZO films were deposited on commercial PET substrates at room temperature by using radio-frequency sputtering with an oxide ceramic target (ZnO:Al2O3, 98%:2%wt)) with a diameter of 75 mm. Before depositing the ZnO:Al thin films, the substrates were cleaned with two method, sample A were cleaned with first method, in this method use Dichloromethane solution followed by a rinse with DI water and dried in nitrogen, and sample B was cleaned with second method that substrates after doing the upper

process before loading in vacuum system the substrates were cleaned in an ultrasonic cleaner with DI Water at 50°C and deride in nitrogen then followed by baking to the chosen temperature (70 °C) during 1 h prior to introduction into the deposition system. The target to substrate distance was kept at 50 mm and the base pressure was $8 \times 10-6$ Torr. The flow rate of the Argon was controlled by mass flow controller (MFC) from10, which corresponds to a working pressure $2.4*10^{-2}$ Torr, and the sputtering RF power was ranged from 100 W to 300 W. The substrate was rotated with speed 1 round per min so that was deposited at low temperature in high power deposition.

The film thickness was measured by a conventional Dektak surface profiler. The film thickness is 100 nm and all samples kept the same thickness in this work. Crystallinity of the films was characterized with an x-ray diffraction system with C (K α) radiation (λ =0.15408 nm) source under an applied voltage of 40 kv and a current of 40 mA. The thickness of the films was determined by a Dektak (Dektak 3) surface profiler. Microstructure and surface morphology of film were observed with a field emission scanning electron microscope. The spectral transmittance of the films was recorded as a function of the wavelength between 100 nm using Cary 500 UV-VIS-NIR spectrophotometer, while the optical parameters of the films such as band gap energy were determined from spectra by a computer program [13,14]. Electrical sheet resistance Rs was determined from the four-point probe method, being the resistivity ρ s obtained using ρ s=Rst, where t is the film thickness estimated from optical characterization.

4. Results and discussion

The crystalline structure of aluminium-doped AZO (AZO:AI) films deposited on PET substrate was examined by XRD measurement. Fig. 1 shows the effect of RF power on the crystal structures of AZO:AI films sputtered at room temperature. The diffraction peak around 34° was observed corresponding to the (002) orientation for all the samples. This phase was confirmed as AZO polycrystalline with wurtzite structure and no other crystal phases were found.



Fig.1: XRD pattern of AZO film deposited at room

Fig. 2 shows the SEM surface microstructure of AZO films deposited at 2.5mTorr with various RF power. In the case of 100 W, pebble-like grains and less dense structure with average grain size of 200 nm were observed. This is due to few energetic particles hit the substrate. Therefore, the mobility of atoms deposited on the substrate is small. As a result, small grains and many grain boundaries are formed. A

densified structure with much larger grains was started to develop as the sputtering power increased to 150 W. As RF power is further increased to 200 W, distinct and independent grain morphology was formed with some ridges along the pyramids. The film exhibits a surface pyramidal morphology. This surface texturing is a consequence of the nucleation of oriented *c*-axis grains that grow geometrically and impinge laterally. Typically, obvious grain boundary characteristics were not observed, although the size of pyramid-like grains has a dimension of 120 nm long and 80 nm width for RF power 250 W. At 300 W RF power, a less dense microstructure and the formation of island-like grains was the result of the columnar growth.



Fig.2: FESEM images of AZO films deposited at (a) 100W, (b) 150W (c) 200W(d) 0.4, (e) 250W (f) 300W

Fig.3 shows the total transmission properties of the AZO films deposited at different RF power ranging from 100 to 300 W at room temperature. All these films demonstrate good optical transmittance (over 80%) in the visible and near infrared spectrum. It is observed from the spectrum that the transmittance of the film reaches its maximum when RF power is at 200W indicating that it is the optimal RF power to prepare the transparent AZO films.



Fig3: Optical spectra for AZO films deposited at different RF power deposited.

Fig.4 shows the effect of sputtering RF power on the electrical resistivity of the AZO films deposited on PET for 30 min. It is found that the electrical resistivity of the films started to decrease and obtained to a minimum value of $1.1 \times 10^{-3} \Omega$ -cm as sputtering RF power increasing to 250W.



Fig.4: The resistivity of AZO thin film deposited on PET substrate.

5. Conclusion

We prepared highly transparent conductive AZO thin films on PET substrate at room temperature by rotation substrate above the target with specified speed at high RF power. The deposition rate increased with the increase of the RF power in both substrates. The Diffraction peak of (002) plane was observed in all the AZO thin films except in AZO thin film deposited on PET substrate at RF power of 250W. AZO thin films deposited on PET substrate at various RF power

have an average transmittance of 80% in the visible range (400–800nm). The resistivity was $1.2 \times 10^{-2} \ \Omega$ -cm at input power of 250W. By employing this method at room temperature, we could obtain the AZO thin film with low resistivity on PET substrate before anneailing, which is a potential use for the flexible display applications.

6. References

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