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Influence of ZnO buffer layer thickness on the electrical and optical properties of indium zinc oxide thin films deposited on PET substrates

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Abstract

The influence of the ZnO buffer layer thickness on the electrical and optical properties of In_2O_3–10 wt.% ZnO and ZnO bilayers deposited on polyethylene terephthalate (PET) substrates by RF magnetron sputtering were investigated. The optimum ZnO buffer layer thickness was found to be 90 nm which gives the lowest electrical resistivity of the bilayer of IZO and ZnO deposited on the PET substrate. The surface roughness decreases and diffusion of moisture and gas is more efficiently restrained, which contributes to lower the resistivity of the bilayer as the ZnO buffer layer thickness is increased. On the other hand, the total resistivity of the bilayer increases as the ZnO buffer layer thickness is increased because the resistivity of ZnO is higher than that of IZO. Introduction of a ZnO buffer layer does not nearly affect the IZO/ZnO/PET sample.

Keywords: Optical properties; Electrical properties; Transparent conducting oxide; ZnO

1. Introduction

Transparent conducting oxides (TCOs) have wide applications in the fields of optoeletronic devices such as organic light emitting diodes (OLEDs), liquid crystal displays (LCDs), electroluminescence devices, and solar cells as transparent electrodes because they have high electrical conductivity and high optical transparency in the visible region. Since flat panel displays (FPDs) such as LCDs and OLEDs particularly tend to be designed toward a large screen with high quality and preciseness, TCO thin films become more and more important in realizing FPDs with high quality. Indium tin oxide (ITO) thin films, which are most widely used as TCOs, must be deposited at a temperature higher than 250 °C and then annealed at a temperature higher than 300 °C in order to obtain high transmittance and conductivity [1]. This high temperature postannealing makes the ITO films rough due to crystallization. The rough surface of the ITO films, in turn, makes the surface of the organic overlayers rough, which results in significant deterioration of the device reliability. For this reason, it is much effort to find TCO materials which will replace ITO that have been made and amorphous IZO (a-IZO) has attracted much attention as an alternative to ITO [2].

Besides the smoother surface, amorphous IZO has several advantages over ITO as follows: (1) a-IZO has higher structural stability at higher temperatures than ITO. The amorphous structure of IZO is maintained up to a temperature as high as 350 °C, which ensures good stability in both electrical and optical properties. (2) a-IZO has higher chemical stability than IZO. In particular for FPD applications TCO films must be prepared at low temperatures below 200 °C to avoid thermal stress on the TFT devices or the flexible substrates. However, ITO films deposited at lower temperatures have lower resistance to moist heat, higher electrical resistivity, lower optical transmittance and poorer chemical stability. (3) a-IZO has the lowest resistivity when no reactive oxygen is added to the sputter chamber which simplifies the deposition process. (4) a-IZO has a lower internal stress which prevents exfoliation from the substrate during device fabrication processes. (5) a-IZO has better wet-etch performance [2–4].

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Deposition of TCO films on glass substrates has been widely investigated during last decade. Recently the necessity of studying deposition of TCO films on polymer substrates has increased because polymer substrates are suitable for flexible displays and electronics the demands of which are expected to increase explosively in the near future. Polymers have merits that they are cheaper and lighter than glass, but they also have several demerits. In comparison with glass substrates, polymer substrates have a lower thermal resistance, a weaker mechanical strength and a higher thermal expansion coefficient (20×10^{-6} K [5]). The difference in thermal coefficient between polymer substrates and ZnO films (4.75×10^{-6} K [6]) may result in residual thermal stress-induced defects. Other short-comings of polymers as substrate materials for TCO are that they easily absorb moisture and gas [7]. Therefore, it is necessary to use a buffer layer when TCO films are deposited on polymer substrates which will make the polymer substrate surface smoother and reduce diffusion of vapor and oxygen. In this work the electrical resistivity and transmittance properties were investigated for the IZO thin films deposited on polyethylene terephthalate (PET) substrates with ZnO layers by using an RF magnetron sputtering technique.

2. Experimental procedure

IZO thin films were deposited on PET substrates by RF magnetron sputtering using a 2 in. IZO (In_2O_3: 90 wt.%, ZnO: 10 wt.%) target. The PET substrate surfaces were cleaned in an ultrasonic cleaner for 10 min with isopropyl alcohol and then blow dry with nitrogen before they were introduced into the sputtering system. Before the deposition of IZO thin films, ZnO buffer layers were deposited using a 2 in. ZnO target. A ZnO buffer layer is expected to reduce the damage, which would be done to the PET surface during deposition of the IZO films without using a buffer layer, and to prevent chemical reactions with oxygen and moisture in the air and their diffusion into the PET substrate. Details of the sputter deposition process parameters for IZO thin films are shown in Table 1. A gas mixture of Ar and O (the flow rate of Ar and O_2 are 20 and 10 sccm, respectively) was blown into the sputter chamber for 30 min to reduce the damage by ion bombardment during sputtering process and to decrease the electrical resistivity of IZO films.

For the prepared samples X-ray diffraction (XRD) analysis was performed to investigate the crystalline structure of the IZO films. An α-step (Dektak-3) was used to measure film thicknesses. The carrier concentrations, carrier mobilities and electrical resistivities of the films were determined by using Hall effect measurement system (HEM-2000). The optical transmittance was made by using an UV–vis spectroscope.

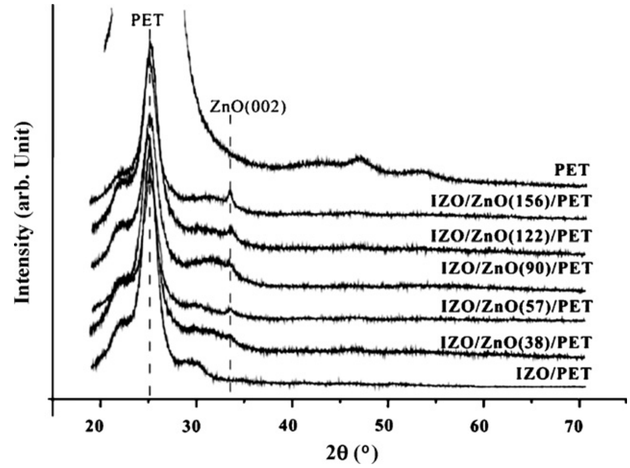

Fig. 1. X-ray diffraction patterns of IZO thin films deposited on the PET substrate with or without a ZnO buffer layer.

3. Results and discussion

Fig. 1 shows the X-ray diffraction spectra for IZO thin films deposited at room temperature on PET substrates with and without ZnO buffer layers with different thicknesses. The highest peak appearing at a 2θ of $\sim$25 °C indicates the PET substrate. A small peak appearing at a 2θ of $\sim 34^{\circ}$ is due to the ZnO buffer layer. No other distinct peak is found, which indicates that the IZO thin film is amorphous as was expected. All as-deposited IZO films are amorphous since the crystallization temperature of IZO films are in a temperature range from 400 to 500 °C.

The carrier concentration, carrier mobility and electrical resistivity of the IZO thin films deposited on PET substrates with or without ZnO buffer layer are plotted as a function of the buffer layer thickness in Fig. 1. The IZO and ZnO films were deposited under the standard process condition shown in Table 1. The IZO/ZnO/glass samples are also shown for the purpose of comparison. Fig. 2 clearly shows that introduction of a ZnO buffer layer with proper thickness increases the carrier concentration and mobility and thus lower the resistivity of the IZO film. The resistivity of the IZO/ZnO/PET sample decreases at 90 nm and then the resistivity increases with the continued increase in the buffer layer thickness. In other words, the optimum resistivity of the IZO film is 90 nm.

Table 1
The process parameters for sputter deposition of IZO and ZnO films

	RF power (W)	Pressure (base pressure)	Substrate temperature	Distance (between target and substrate) (cm)	Substrate	Thickness (nm)	Ar/O_2 gas flow ratio (sccm)
ZnO buffer film	80	4.3×10^{-3} Torr (1×10^{-7} Torr)	RT	8	Glass, PET	0, 38, 57, 90, 122, 156	20:10
IZO film					ZnO/Glass, ZnO/PET	300	30:0

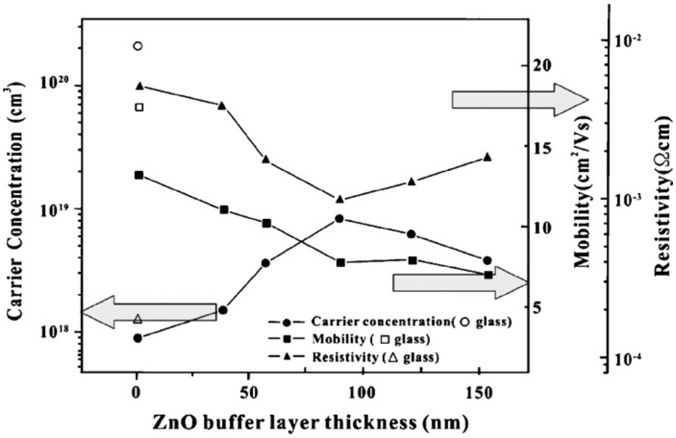

Fig. 2. Dependence of the carrier concentration, carrier mobility and resistivity of IZO/ZnO thin films on the ZnO buffer layer thickness.

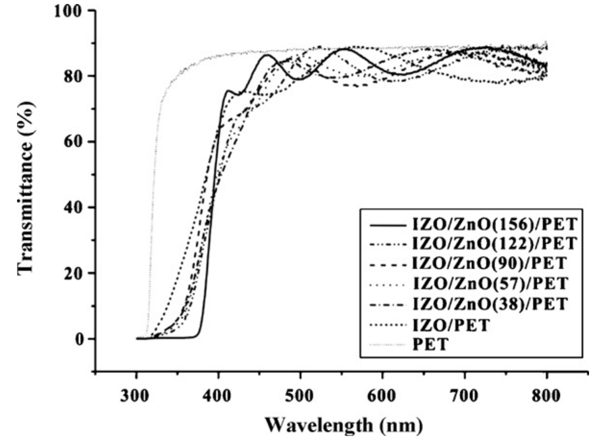

Fig. 5. Transmission spectra of IZO/ZnO/PET samples with different ZnO layer thicknesses.

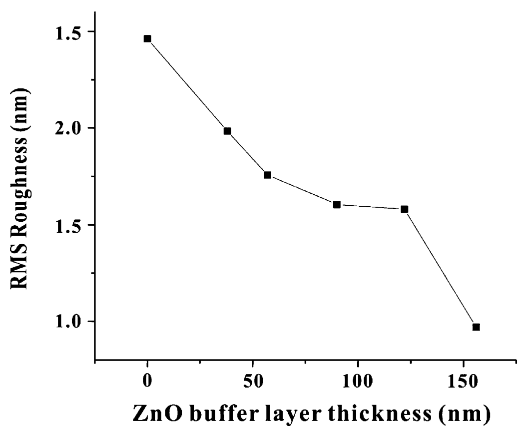

Fig. 3. Dependence of the IZO film surface roughness on ZnO buffer layer thickness.

The effective resistivity ρ of a bilayer structure consisting of an IZO layer with a thickness of t_{IZO} and a ZnO layer with a thickness of t_{ZnO} can be expressed as

$$\frac{1}{\rho} = \frac{1}{R_T t_T} = \frac{1}{R_{IZO} t_{IZO}} + \frac{1}{R_{ZnO} t_{ZnO}} \quad (1)$$

where R_{IZO} and R_{ZnO} are the sheet resistances of an IZO layer and a ZnO layer, respectively, t_{IZO} and t_{ZnO} are the thicknesses of the two layers and $t_T = t_{IZO} + t_{ZnO}$. Therefore, the total resistance increases as the ZnO buffer layer thickness is increased, which is a negative effect, because the resistivity of ZnO is higher than that of IZO.

On the other hand, the IZO film surface becomes smoother as shown in Figs. 3 and 4 as the buffer layer thickness increases. Diffusion of moisture and gas from the PET substrate to IZO film is restrained more effectively as the buffer layer thickness is increased. The smoother surface and restraint of diffusion of moisture and gas with an increase in the buffer layer thickness is a positive effect. Therefore, it can be said that an optimum buffer layer thickness exists when a minimum resistivity is obtained. The optimum buffer layer thickness determined by a trade-off between the positive and negative effects discussed above.

The optical transmittance spectra of the IZO/ZnO/PET samples in the wavelength range from 300 to 800 nm are shown in Fig. 5. The transmittance spectra of the PET substrate and the IZO film are also shown for the purpose of comparison and they have optical transmittance of ~86 and ~80%, respectively. The overall transmittance of the IZO/ZnO/PET sample is almost the same as the IZO/PET sample. The transmittance of the IZO/ZnO/PET sample seems to be almost independent of the buffer layer thickness. The IZO films with a ZnO buffer layer have higher transmittance in the red and blue regions but lower transmittance in the yellow region. This dependence of the transmittance of the IZO/ZnO/PET samples on the wavelength can be explained with etalon interference effect, which can be inferred by changing color for different view angles.

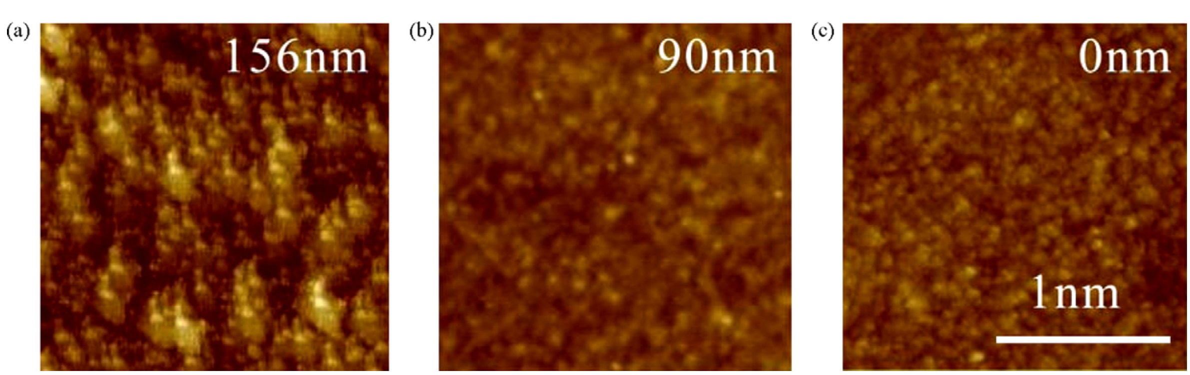

Fig. 4. AFM images of IZO film surface with different ZnO buffer layer thicknesses: (a) 156 nm, (b) 90 nm and (c) 0 nm.

4. Conclusions

The optimum ZnO buffer layer thickness was found to be 90 nm which gives the lowest electrical resistivity of the bilayer of IZO and ZnO deposited on the PET substrate. The surface roughness decreases and diffusion of moisture and gas is more efficiently restrained, which contributes to lower the resistivity of the bilayer as the ZnO buffer layer thickness is increased. On the other hand, the total resistivity of the bilayer increases as the ZnO buffer layer thickness is increased because the resistivity of ZnO is higher than that of IZO. Introduction of a ZnO buffer layer does not nearly affect the IZO/ZnO/PET sample.

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