Electrical and Optical Properties of ITO Films Sputtered by RF-bias Voltage and In-Sn Alloy Target

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ITO thin films were deposited on PET and soda-lime glass substrates by a dc reactive magnetron sputtering of In-Sn alloy metal target without substrate heater and post-deposition thermal treatment. The dependency of rf-bias voltage and substrate power during deposition processing was investigated to control the electrical and optical properties of ITO films. The range of rf bias voltage is from 0 to -80 V and the substrate power is applied from 10 to 50 W. The minimum resistivity of ITO film is 5.4×10^{-4} Ωcm at 50 W power and rf-bias voltage of –20 V. The best transmittance of ITO films at 550 nm wavelength is 91 % in the substrate power of 30 W and rf-bias voltage of –80 V.

Keywords : Indium tin oxide (ITO), RF-bias voltage, Substrate power; In-Sn alloy target, Magnetron sputtering

1. INTRODUCTION

Transparent conducting indium tin oxide (TC-ITO) films have high transparent properties in the visible wavelength region and a low electrical resistivity. Therefore, they have been widely used in many applications such as transparent electrodes, antireflection coating, heat reflecting mirrors, transparent electro-magnetic shield coating, display devices, and opto-electric and photovoltaic devices[1,2]. Recently, TC-ITO films with excellent quality have become necessary to adapt the technological demands for flat panel displays. Over the years different methods of preparing ITO films have been reported, such as electron beam evaporation (EB), chemical vapor deposition (CVD), magnetron sputtering (MS), electron cyclotron resonance (ECR) plasma sputtering, and spray pyrolysis (SP)[3-5]. However the deposition method has been established completely to fabricate ITO films that accommodate recent demands. The magnetron sputtering method with a metal alloy target has been widely used to get highly conducting and transparent ITO films with board uniformity and excellent adhesion to the substrates. The electrical and optical properties of ITO films strongly depend on the sputtering conditions. Therefore, most of the reports are devoted to controlling the sputtering conditions and heat treatments to obtain high trans-parency and conductivity. Bender, et al.[3] showed the sputtering method using the rf-superimposed dc magne-tron gas discharge of 50 % rf power and 50 % dc power. Also it was reported by Futagami, et al.[5] that the properties of ITO films were studied by the effect of rf-enhanced dc sputtering using a one-turn coil above target.

In the magnetron sputtering, it is well known that the energetic particles such as negative Ar ions have some impact on the ITO film. Honda, et al.[6] reported that the composition of oxide film was mainly dependent on substrate bias, and proper ion impact on the deposited film caused improvement of film properties. So we installed the magnetron sputtering apparatus to be able to apply rf-bias voltage to control the energetic particles and investigated the effect of energetic particles on the electrical and optical properties. In general, the properties of ITO films exhibit a strong dependence of temperature in the sputtering process. It has been recently required that transparent conducting ITO (TC-ITO) films are prepared on several polymers as a substrate material. However, the preparation of ITO thin films using polymer substrates should be performed possibly without heat treatment, so that the coating methods and deposition parameters of plasma processing are greatly important.

In this paper, ITO films were deposited on flexible transparent substrate by a magnetron sputtering with In-Sn alloy target. As the above mentioned, the energetic particles such as negative ions and secondary electrons caused some damage on film surface during the sputtering deposition. In order to control the energetic particles, an effect of rf-bias voltage for the improvement of film properties was investigated. By a relatively lower rf-bias voltage of –80~0 V, it was possible to obtain the excellent TC-ITO films with a low resistivity and high transmittance in the visible range. Therefore, both parameters such as sputtering power and re-bias voltage in various deposition conditions are selected in this research. For the TC-ITO film properties, the growth rate, electrical resistivity, carrier concentration, Hall mobility, and optical transmittance were measured.

2. EXPERIMENTAL

ITO thin films were deposited on polyethylene terephthalate (PET) and soda-lime glass substrates by a dc reactive magnetron sputtering (Leybold, AG-L560) of In-Sn alloy metal target. The composition of alloy metal target was 90 wt% In-10 wt% Sn producted by Cerac, and the size of a disk-type target was 2 inch diameter and 1/4 inch thick. All substrates were ultrasonically cleaned for 5 min using methanol as a solvent, and blown dry in N_2 gas. The substrate temperature was not applied during the film deposition, and subsequently post-annealing treatment for as-deposited ITO films was not performed.

The deposition parameters such as dc power and rf-bias voltage were examined. The substrate was applied negative rf-bias voltage from 0 to -80 V. Of course, the oxygen concentration in ITO film has been one of the important factors, which influence the film properties. Oxygen vacancy produces carriers that contribute to the resistivity. Therefore, we have reported to estimate the relationship between partial oxygen pressure and film properties[7]. The reactive sputtering gases of high purity oxygen (99.999 %) and argon (99.999 %) were introduced into the chamber. The low dc power from 0 to 50 W was applied to eliminate the deformation of polymeric substrates caused by the concomitant temperature effect. A serious problem in the reactive sputtering of metallic alloy target is the oxidation of metal target surface, well known as a poisoning phenomenon. Therefore, the target surface should be cleaned necessarily before every sputtering run. Moreover, the target was pre-sputtered in argon atmosphere for about 10 min. in order to remove the oxidizing surface and impurity layer, which may have formed during exposure to air. Other sputtering condi-tions are as follows; target-to-substrate distance of 4.5 cm, working pressure of 5 mTorr, oxygen partial pressure of 8~13 %, and substrate rotation speed of 10 rpm.

Film thickness measurements were carried out using a α-step 500 surface profilometer manufactured by Tensor. Optical transmittance measurements were performed using a UNICAM 8700 spectrophotometer made by Phillips, in the visible and near-infrared range of 200 nm-22 μm. Electrical resistivity and sheet resistance measurements were measured with a standard 4-point probe (CMT-SR 1000, Chang-Min Co.) and Hall measurement system (Lake-shore EMA-CS electromagnet system and Keithley measurement system with 7065 Hall Card).

3. RESULTS AND DISCUSSION

Figure 1 shows the growth rates of ITO film on PET and glass substrates with increasing rf-bias voltage. The growth rate in sputtering power of 10~50 W generally seems to decrease with increasing rf-bias voltage, and then saturate over rf-bias voltage of –60 V. In case of applying rf-bias voltage, it seems that the reducing trend of growth rate is due to a kind of etching effect by high bombardment of sputtered particles, energetic Ar neutrals and negative oxygen ions with high energy.

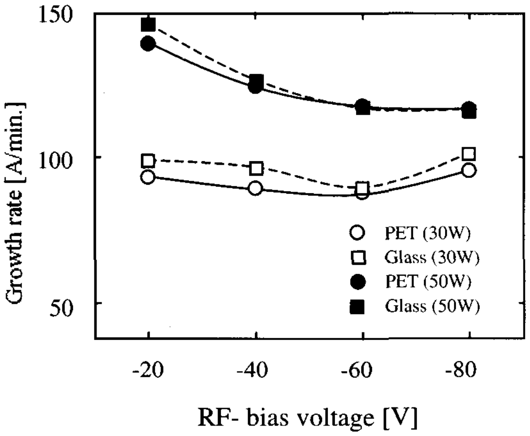


Fig. 1. Growth rates of ITO film with rf-bias voltage at sputtering power of 30 and 50 W.

Figure 2 shows the growth rate of ITO films on PET and glass substrates with an increase of sputtering power. The growth rate in power of 50 W increases about 3 times higher than that of 10 W. As shown in figure, the growth rate increases with an increase of sputtering power. It is clear that the width of plasma region increases with increasing sputtering power. This suggests that the virtual source is shifting away from the target with increasing sputtering power[8]. The increase of sputtering power increases the self-bias at the target, which causes an increase in the density of sputtered neutrals and their average energy.

The electrical resistivity of ITO films at oxygen partial pressure of 8 % as a function of rf bias voltage are represented in Fig. 3. The resistivity of ITO film decreases slightly with increasing rf-bias voltage, and then shows the increasing trends in all sputtering power. But the resistivity in sputtering power of 50 W increases gradually with an increase of rf-bias voltage. The electrical resistivity mainly depends on the carrier concentration. We found the trend that the carrier concentration of ITO film increases initially until the maximum and then decreases with increasing rf-bias voltage[7]. The carrier concentration increases with increasing oxygen deficiency, and therefore the resistivity is decreased.

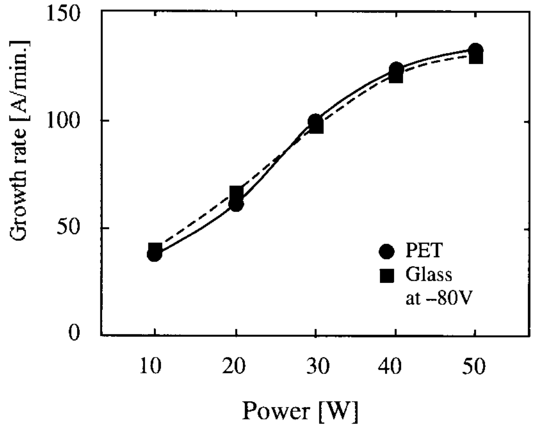


Fig. 2. Growth rates of ITO film on PET and glass substrates as a function of sputtering power.

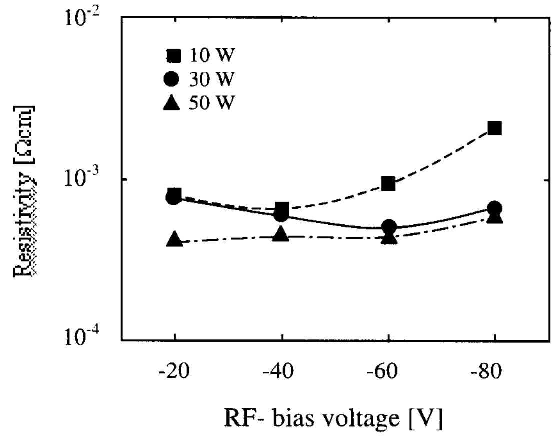


Fig. 3. Electrical resistivity of ITO films grown on PET as a function of rf-bias voltage.

The carrier concentration, mobility, and electrical resistivity of ITO films at oxygen partial pressure of 10 % and rf-bias voltage of –80 V as a function of sputtering power are represented in Fig. 4. The resistivity reduces abruptly with increasing power, and saturates gradually above 30 W. On the other hand, the carrier concentration increases quickly, and then saturates. The resistivity of ITO film is strongly related to the carrier concentration as discussed already in Fig. 3. As shown in figure, it seems that the resistivity is not caused clearly by the mobility. The mobility increases from 11 cm^2/Vs to 28 cm^2/Vs with increasing substrate power, and shows a reducing trend at 50 W. It seems that the change in orientation of ITO films with a variation of power may cause the change in mobility. The reported mobilities at room temperature were about 15~40 cm^2/Vs, and the highest mobility was approximately 160 cm^2/Vs for In_2O_3 single crystal[9]. The mobility of ITO film is strongly influenced by disorder because of the particular structure of In_2O_3. Consequently, there is a scattering effect on carriers. Thereby the mobility is relatively lowered. The scattering may influence the electrical and optical properties of ITO films.

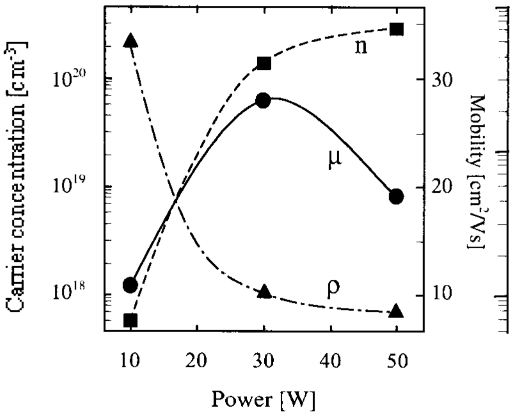
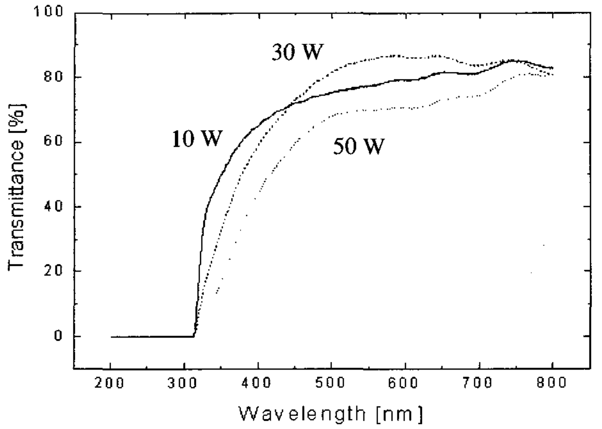
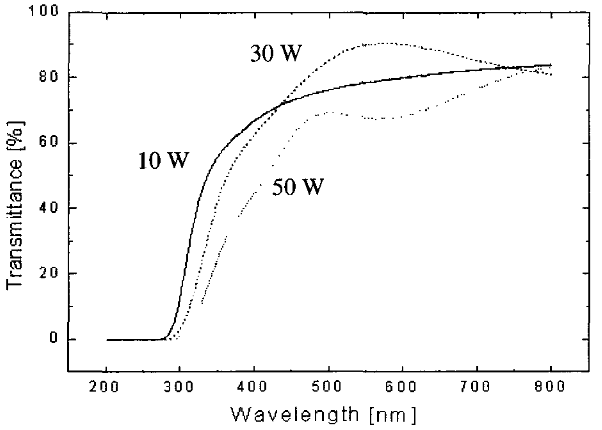


Fig. 4. Carrier concentration, mobility, and resistivity of ITO films on PET at rf-bias voltage of –80 V as a function of sputtering power.

Figure 5 shows the optical transmittance of ITO films deposited on PET and glass substrates at rf-bias voltage of –80 V with different sputtering power. The transmittance patterns of ITO films on both substrates are similar, and the increase of power shows the lowering of average transmittance in 300~800 nm range. Terzini et al. reported the similar tendency of lowering transmittance with increasing power[10]. However, the transmittance of 30 W produces the highest value of 91 % at 550 nm wavelength.



(a) PET



(b) Glass

Fig. 5. Optical transmittance of ITO films on PET and glass substrates at rf-bias voltage of –80 V with different sputtering power.

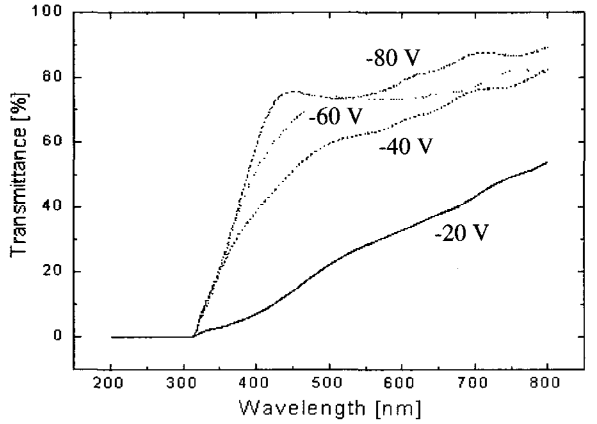


Fig. 6. Optical transmittance of ITO films on PET substrate at power of 50 W as a function of rf-bias voltage.

Figure 6 presents the optical property of transmittance in the visible range of 300~800 nm as a function of rf-bias voltage. The patterns of average transmittance increase with increasing rf-bias voltage. It was found that the transmittance showed the same tendency as oxygen content in ITO films[11].

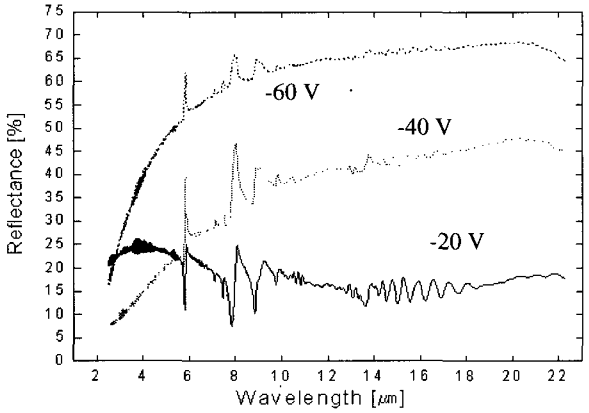


Fig. 7. Reflectance spectra of ITO films with different rf-bias voltage.

The reflectance spectra of ITO films deposited at various rf-bias voltages are given in Fig. 7. The optical reflectance was measured in the infrared wavelength range of 2~22 μm. It should be noted that the reflectance in the infrared region increases significantly as the rf-bias is raised. From the figure, the ITO films at rf-bias voltage of –60 V are potential candidates for applications in infrared mirrors, since the reflectance is high at wavelengths greater than 2 μm.

4. CONCLUSIONS

ITO thin films were deposited on PET and soda-lime glass substrates by a dc reactive magnetron sputtering of In-Sn alloy metal target. The substrate power and rf substrate bias in the deposition conditions are important factors during ITO films processes. The results are summarized as follows.

(1) The growth rate in power of 10~50 W shows to decrease with increasing rf-bias voltage, and then saturate over rf-bias voltage of –60 V. The growth rate increases with an increase of sputtering power.

(2) The resistivity of ITO film decreases slightly with increasing rf-bias voltage, and then shows the increasing trends in all sputtering power. The minimum resistivity of ITO film is 5.4×10^{-4} Ω cm.

(3) The resistivity reduces abruptly with increasing power, and however the carrier concentration increases quickly. The mobility increases with

increasing substrate power, and shows a reducing trend at 50 W.

(4) The increase of power shows the lowering of average transmittance in 300~800 nm range. The patterns of average transmittance increase with increasing rf-bias voltage. The best transmittance of ITO films at 550 nm wavelength is 91% in the substrate power of 30 W.

(5) The reflectance in the infrared region increases significantly as the rf-bias is increased.

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