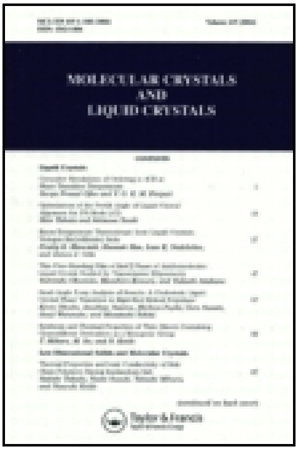

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In this study, indium tin oxide (ITO) films were deposited at 300°C on glass substrates by low-frequency (60 Hz) magnetron sputtering technique. The influence of oxygen flow rate on the structural, electrical and optical properties of ITO thin films was investigated. The oxygen flow rate was varied from 0 to 8 sccm. We obtained the most superior property of ITO films at oxygen flow rate of 0.5 sccm. The films are found to show preferential orientation both (222) and (400) planes in the XRD patterns. The resistivity of the films with the thickness of 140 nm is 2.1×10^{-4} *ohm·cm with 88% optical average transmittance in visible range (500–800 nm). Therefore, we conclude that transparent conducting ITO films prepared by low-frequency magnetron sputtering can be applied for various flat panel displays.*

Keywords: flat panel displays; indium tin oxide films; magnetron sputtering; oxygen flow rate; resistivity; transparent conducting oxide films

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INTRODUCTION

Recently, transparent conducting oxide (TCO) films have been widely used as transparent electrodes for solar cells, touch panels, flat liquid crystal displays, optoelectronics, organic light emitting diode and so on. In particular, indium tin oxide (ITO) films have been preciously used because of high optical transmittance in the visible region, high electrical conductivity and processing compatibility [1–3].

Generally, ITO films are deposited by the radio frequency (13.56 MHz) magnetron sputtering, direct current magnetron sputtering, chemical vapor deposition, spray pyrolysis, laser ablation, and electron beam method [4–6]. Although many methods can be used to deposit ITO films, more attention has been paid to sputtering method because of the easy control of the deposition parameters. In particular, reactive magnetron sputtering is one of the most important sputtering methods and has been widely used for depositing ITO films. In this process, the oxygen gas is usually used as reactive gas. So the oxygen flow rate is a very important parameter in reactive sputtering process [7]. We have tried to deposit the films by low-frequency (LF) method to obtain the high quality films, based on the fact that the LF (60 Hz) plasma has peculiar properties such as non-continuous discharge, relatively high electron temperature, and small sample damage [8,9].

In this work, the influence of oxygen flow rates on the structural, electrical and optical properties of ITO films prepared by LF magnetron sputtering method has been reported. We interpreted the results in terms of the variation of film properties depending on the oxygen flow rate.

EXPERIMENTAL

ITO films were deposited on glass substrate at 300°C by low-frequency (60 Hz) magnetron sputtering method. The schematic diagram of experimental setup for a LF magnetron sputtering system was shown in Figure 1. Indium-tin alloy (9:1) of 99.99% purity was used as a target source. The size of the target was 3 inches in diameter and 5 mm in thickness. The glass was used as the substrate and the samples were of $50 \times 50\,mm^2$. The distance between the target and the substrate was about 100 mm.

The vacuum chamber was evacuated down to a pressure of 5×10^{-6} Torr prior to sputtering. After evacuation, a mixed gas of argon (99.999%) and oxygen (99.999%) was introduced into the chamber. The flow rates of oxygen reactive gas were varied from 0 to 8 sccm. Before sputtering, the target was pre-sputtered under biasing of

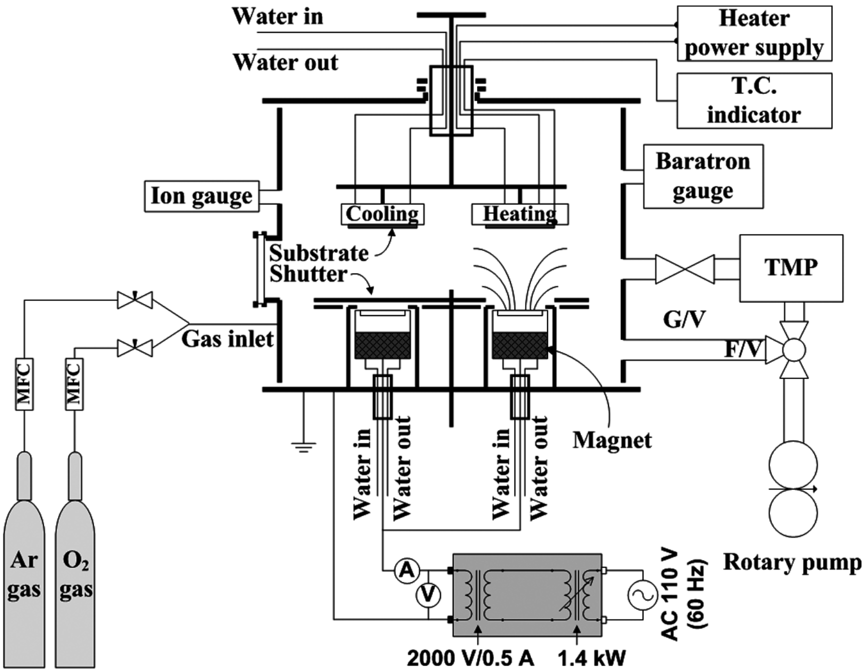


FIGURE 1 LF (60 Hz) magnetron sputtering system.

320 V for 1 minute, keeping the target covered with a shutter, in order to remove the surface oxide layer. The detailed sputtering conditions of ITO films are summarized in Table 1.

We analyzed structural, electrical, and optical properties of ITO films prepared on glass substrates. The thickness of films was

TABLE 1 Summary of Sample Preparation Conditions

Sample no.	No. 1	No. 2	No. 3	No. 4	No. 5	No. 6	No. 7
T-S distance [mm]				100			
LF power voltage [V]				320			
Base pressure [Torr]				5×10^{-6}			
Argon flow rate [sccm]				30			
Oxygen flow rate [sccm]	0	0.5	1	2	3	5	8
Sputtering time [min.]				15			
Substrate temp. [°C]				300			
Substrate				Sodalime glass			
Film thickness [nm]	133	140	146	177	182	165	168

determined using alpha-step (Veeco, Dektak 3) and SEM (Hitachi S-4200). The crystal structure and resistivity of the films was investigated using XRD (Mx Labo) and 4-point probe (Mitsubishi, MCP-T360), respectively. The carrier concentration and mobility were measured using Hall effect measurement system (EGK, HEM-2000). The optical transmittance and surface morphology were measured UV-Visible spectrophotometer (Shimadzu, UV-1601PC) and AFM (Digital Instrument, Nanoscope IIIa), respectively.

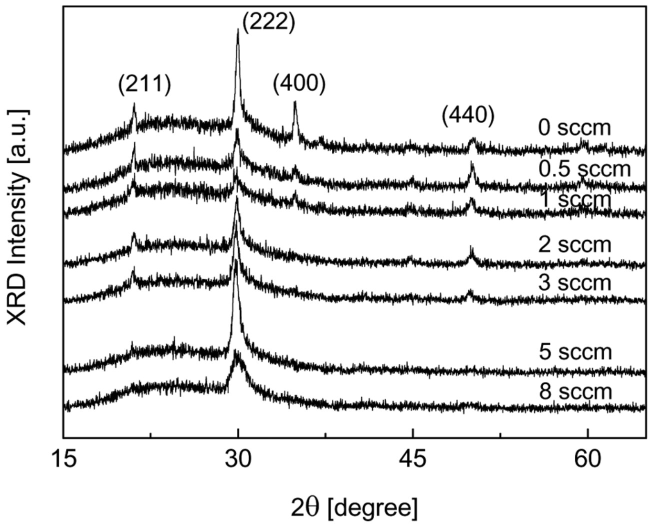
RESULTS AND DISCUSSION

Figure 2(a) shows the XRD patterns of ITO films deposited at various oxygen flow rates within the range 0–8 sccm. The sputtering conditions were 320 V (LF, 60 Hz), 30 sccm of argon flow rate, and substrate temperature of 300°C. Mainly observed XRD peaks in ITO films are (222) and (400) preferred orientation, representing that the structure of ITO films is a body-centered cubic structure. In argon alone, there are two dominant planes in the film, (222) and (400). As the oxygen flow rate is increased, the (400) disappears and (222) peak becomes the dominant plane.

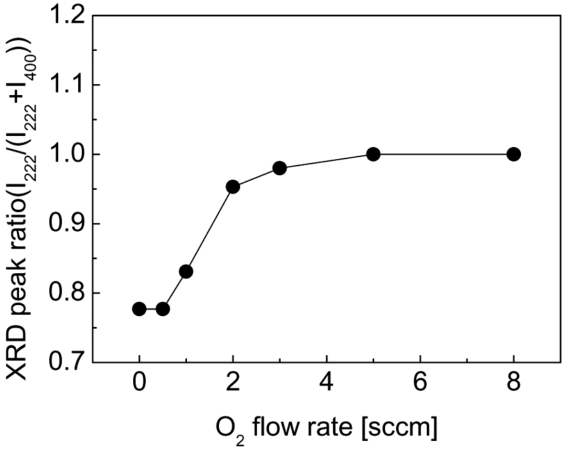
In order to get some detailed information about the film structure, the fitting of the XRD peak has been done for these films. The ratio of XRD peak intensities (I_{222}/I_{400}) as a function of the oxygen flow rate are shown in the Figure 2(b). The change in preferred orientation can be explained as the result of Jun *et al.* [10]. They explained that the driving forces of this change in preferred orientation are the concentration of oxygen vacancies and their subsequent diffusion.

The oxygen vacancies play an important role in atomic diffusion and the atoms can diffuse through these vacancies. Therefore, as the oxygen flow rate is increased, the concentration of oxygen vacancies is decreased. As a result, the (222) peak will dominate in higher oxygen content in the sputtering ambient since the close packed plane is (222) in the body centered cubic In_2O_3 crystal. It should be noted the preferred orientation of ITO films on the glass substrates was closely related to the mobility and grain size [3].

The electrical characteristics of ITO films also showed dependence on the oxygen flow rate. Figure 3 shows the resistivity of ITO films deposited at various oxygen flow rates on glass substrates. It can be seen that the resistivity remains low 2.1×10^{-4} ohm·cm for the oxygen flow rates ≤ 0.5 sccm and then increases sharply. The resistivity of the films is attributed to the combined effect of changes in carrier concentration and Hall mobility.



(a)



(b)

FIGURE 2 (a) The XRD patterns of ITO films deposited at various oxygen flow rates on glass substrates and (b) the ratio of the XRD peak intensities (I_{222}/I_{400}) as a function of the oxygen flow rate (300°C, argon 30 sccm).

The Hall mobility and carrier concentration of ITO films with oxygen flow rates are shown in the Figure 4. The carrier concentration of the ITO films decrease gradually and Hall mobility increases initially and then decreases as the oxygen flow rate. This trend is

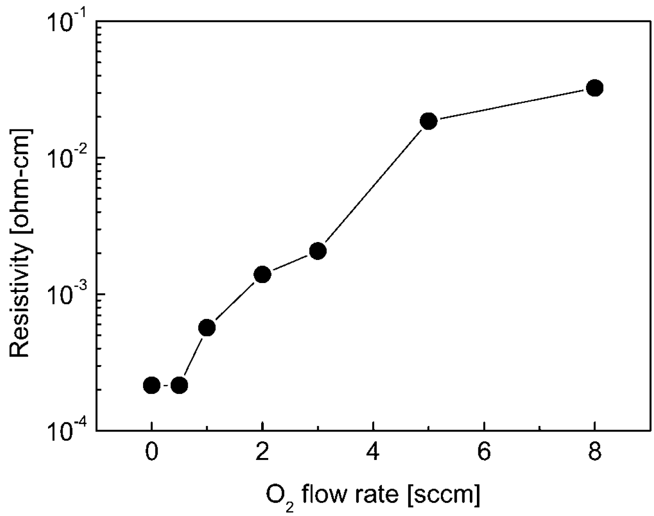


FIGURE 3 The resistivity of ITO films as a function of oxygen flow rates.

similar to the results reported in the earlier studies [11]. The incorporation of oxygen leads to a decrease in oxygen vacancies in the films and hence to a fall in the carrier concentration. The carrier mobility is affected by the crystal structure [12]. A detailed study of this relationship between the carrier mobility and the carrier concentration will be investigated and reported later.

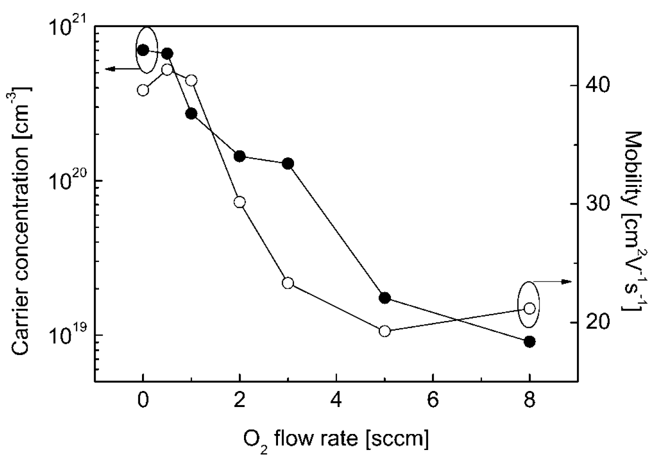


FIGURE 4 The carrier concentration and mobility as a function of oxygen flow rates.

Figure 5(a) shows the optical transmittance spectra of the ITO films deposited at various oxygen flow rates (film thickness) on glass substrates. The optical transmittances of the films at the UV region from 250 to 350 nm were found to behave the gradual red-shift with the

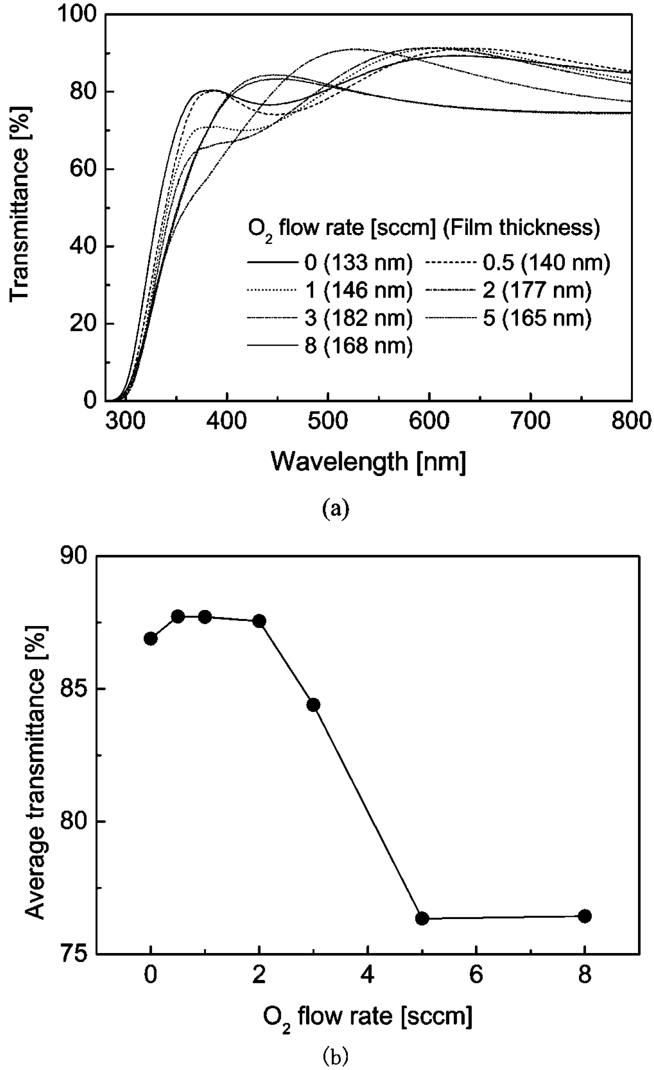


FIGURE 5 (a) The optical transmittance spectra of ITO films deposited at various oxygen flow rates (film thickness). (b) Average transmittance in the visible region (500–800 nm) of ITO films as a function of the oxygen flow rates.

increase in the film thickness. The trends of the gradual red-shift in the optical transmittance are closely related to the bandgap energy of the ITO film [13]. A detailed study of this relationship between the film thickness and the optical transmittance will be investigated and reported later. The variation of the average transmittance in the visible region (500–800 nm) with the oxygen flow rates is shown in Figure 5(b). The average transmittance of the films was slowly saturated with oxygen flow rate up to 2 sccm, and then it rapidly decreased. The average transmittance reaches to a maximum value (88%) at the oxygen flow rate of 0.5 sccm. Thus, the optimum condition of the oxygen flow rate is 0.5 sccm (film thickness 140 nm) from the resistivity and average transmittance.

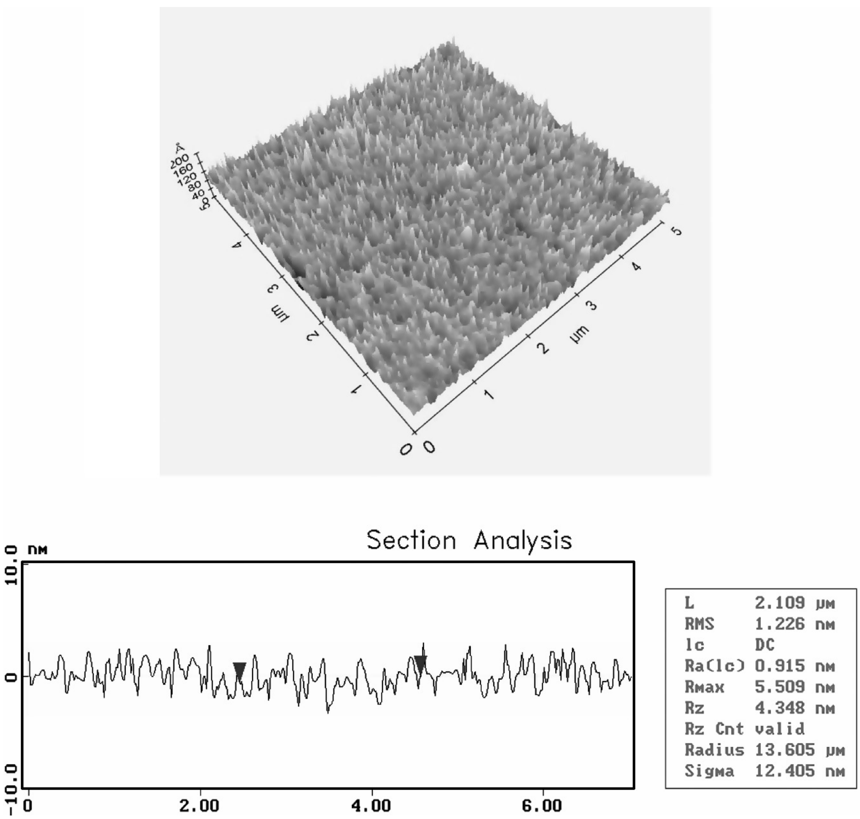


FIGURE 6 The surface morphology of ITO films deposited at the oxygen flow rate of 0.5 sccm (300°C, argon 30 sccm).

The surface morphology of the ITO films prepared at oxygen flow rate of 0.5 sccm for specimens deposited at 300°C is shown in Figure 6. From the figure, the surface morphology of the films seems to be very smooth. The value of surface roughness (root-mean-square) was about 1.2 nm.

CONCLUSIONS

In this study, ITO films were deposited at 300°C on glass substrates using a LF (60 Hz) magnetron sputtering system and influence of oxygen flow rate on the structural, electrical and optical properties of films was investigated. XRD analysis showed that grown film consists of mainly (222) and (400) aligned grains and changes grain alignment with oxygen flow rates. Considering both the optical transmittance and the resistivity, the optimum oxygen flow rate should be 0.5 sccm (film thickness 140 nm) for this sputtering system. ITO films prepared at that flow rate has about 88% optical transmittance in the visible region (500–800 nm) and 2.1×10^{-4} ohm·cm electrical resistivity. As a result, we claim that transparent conducting ITO films prepared by low-frequency magnetron sputtering can be applied for organic light emitting devices (OLEDs).

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