Effects of thickness on the properties of ITO films deposited by a low-frequency magnetron sputtering

Sung Ho Lee, Sang Kooun Jung*, Young June Hur**, Do Kyung Lee, Duck Kyu Park*, Lee Soon Park and Sang Ho Sohn****

Phone : +82-54-450-6498 , E-mail : lpplsh@naver.com

Gumi Institute of Electronics Technology, Gumi, Gyeongsangbuk-do 730-030, Korea

***Department of Physics, Kyungpook National University, Daegu 702-701, Korea**

****Advanced Display Manufacturing Research Center, Kyungpook National University, Daegu 702-701, Korea**

*****Department of Polymer Science, Kyungpook National University, Daegu 702-701, Korea**

Abstract

Indium-tin oxide (ITO) films were deposited by a low-frequency (LF, 100 Hz) magnetron sputtering on glass substrates at the room temperature. The effects of the film thickness on the structural, electrical and optical properties of ITO films were investigated. With the film thickness the films reveal better crystallinity, showing both (222) and (400) planes in the XRD pattern. The optical transmittance and the sheet resistance of the films decreased with the increasing thickness. In addition, the electrical properties of ITO films were improved after annealing in a vacuum.

1. Introduction

Recently, transparent conducting oxide (TCO) films have been widely used as transparent electrodes for touch panels, flat liquid crystal displays, electroluminescent devices, solar cells, antistatic coatings 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 [1-4]. Generally, ITO films have been deposited by the magnetron sputtering deposition method which uses a glow discharge plasma occurring by help of a high frequency (13.56 MHz) or a direct current (DC) power source. We have tried to deposite the films by a low-frequency (LF) method to obtain the high quality films, based on the fact that the LF (100 Hz) plasma has peculiar properties such as non-continuous discharge, relatively high electron temperature, and small sample damage [5].

In this work, we investigated the effects of film thickness on the growth and the structural, electrical and optical properties of ITO films deposited by LF magnetron sputtering. We interpreted the results in terms of the variation of film properties depending on the film thickness.

2. Experimental details

ITO films were deposited on glass substrates by a LF (100 Hz) magnetron sputtering method at the room temperature (RT). 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 0.2 inch in thickness. The glass was used as the substrate and the samples were of 50×50 mm^2. The distance between the target and the substrate was about 100 mm. The vacuum chamber was evacuated down to a pressure of 1×10^{-5} Torr prior to sputtering. The Ar gas was used as a working gas and Ar plasma was generated by a LF power source. Before sputtering, the target was pre-sputtered under biasing of 280V 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. Rightly after sputtering, the as-deposited ITO films suffered a heat treatment at 300 ^{o}C in a vacuum for 10 minutes.

We analyzed structural, electrical, and optical properties of ITO films. The thickness of films was determined using SEM (Hitachi S-4200). The crystallinity of the films was investigated by XRD measurements. The sheet resistance of the films was measured by a 4-point probe (Mitsubishi, MCP-T360). The optical transmittance and surface morphology of the films were also investigated using UV-VIS spectrometer (Shimadzu, UV-1601PC) and AFM (Digital Instrument, Nanoscope IIIa), respectively.

Table 1. The sputtering conditions of ITO films.

Sputter parameters	Ranges
T-S distance [mm]	100
LF Power voltage [V]	280
Frequency [Hz]	100
Base pressure [Torr]	1×10^{-5}
Working pressure [Torr]	3×10^{-3}
Sputtering time [min.]	30, 60, 90, 120
Substrate temperature [°C]	Room temperature

3. Results and discussion

Fig. 1 shows the XRD spectra of ITO films deposited at several sputtering time (i.e. thickness) within the range 30-120 minutes at RT on glass substrates. The ITO film with the lowest thickness (85 nm) was almost in the amorphous phase. However, as the thickness increased, the films showed better crystallinity with (222) preferred orientation mixed with small amounts of (400), (440) and (622). This result suggests that these films begin to crystallize at/above 102 nm in the film thickness. Mainly observed XRD peaks in the ITO films have (222) and (400) preferred orientations, representing that the structures of the ITO films are a body-centered cubic structures [6]. This fact is known to be related to the deposition technique and/or the film thickness [7].

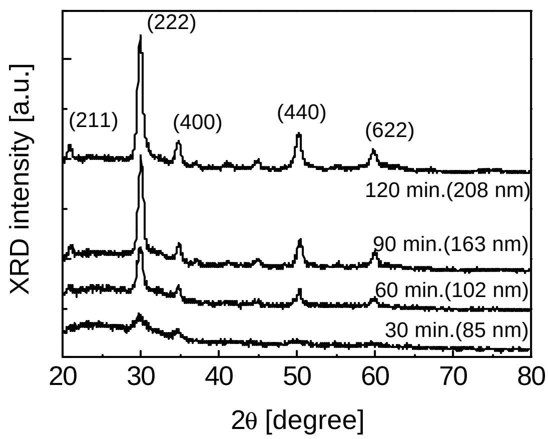


Fig. 1. XRD spectra of the ITO films deposited at several sputtering time (i.e. thickness) on glass substrates. All samples were deposited at RT.

Fig. 2 represents the total crystallinity defined by summing the XRD peaks ($I_{222}+I_{400}$) and the ratio of XRD peak intensities (I_{222}/I_{400}) as a function of the sputter time. The XRD intensity implying the total crystallinity ($I_{222}+I_{400}$) of ITO films increased gradually with the increasing film thickness. It is found that (I_{222}/I_{400}) ratio increases up to 90 minutes with the sputtering time, above which it saturates slowly. This indicates that the preferred orientation along the (400) plane at the higher thicknesses of ITO films increase. It should be noted the preferred orientation of ITO films on the glass substrates was closely related to the carrier mobility and grain size [8].

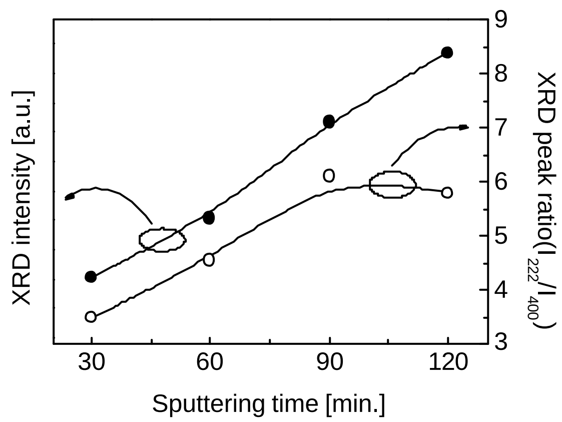


Fig. 2. The variation of the total crystallinity defined by summing the XRD peaks ($I_{222}+I_{400}$) and the ratio of the XRD peak intensities (I_{222}/I_{400}) as a function of the sputtering time.

Fig. 3(a) shows the optical transmittance spectra of the ITO films sputtered for 30, 60, 90 and 120 minutes. 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 increase in the film thickness. The trends of the gradual red-shift in the optical transmittance are closely related to the band-gap energy of the ITO film [5]. Fig. 3(b) shows the variation of the average transmittance in the visible region (400~700 nm) with the sputtering time. The average transmittance was slowly saturated with sputtering time up to 90 minutes, and then it rapidly decreased.

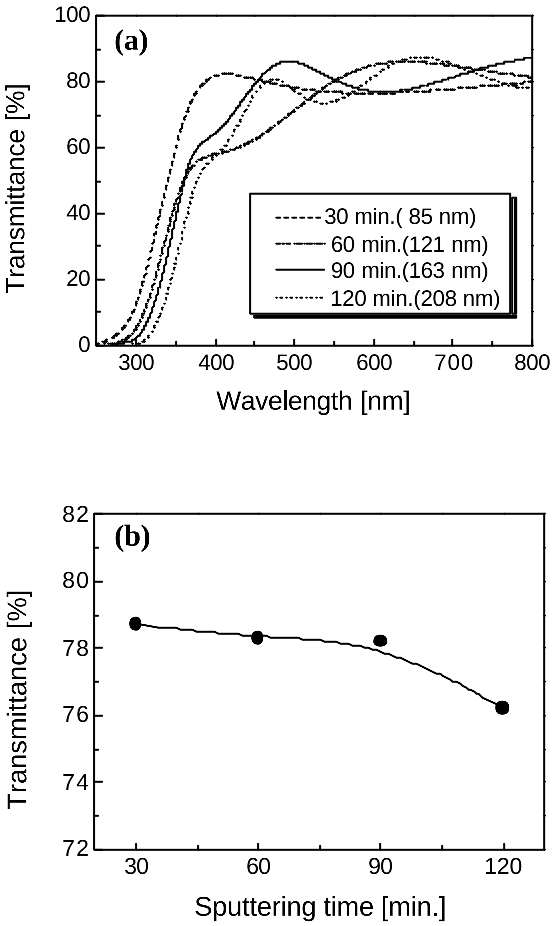


Fig. 3. (a) The optical transmittance spectra of the ITO films deposited at several sputtering time. (b) Average transmittance in the visible region (400~700 nm) of the ITO film as a function of the sputtering time.

Fig. 4 shows the sheet resistance of ITO films having different sputtering time, before and after the annealing at 300 °C in vacuum for about 10 minutes. The sheet resistance of the films decreased with an increase of the sputtering time. The decreases in the sheet resistance are attributed to the increase in the carrier concentration through the film thickness [8]. In addition, the sheet resistance of the annealed films is slightly lower than that of the as-deposited films. The difference is larger in the thinner films. But, over 90 minutes (thickness of 163 nm), the difference is small. Therefore, the annealing effect in the sheet resistance appears more strongly in thinner films. This trend is similar to the results reported in the earlier studies [9-10]. Annealing effect has also influence on the surface morphology of films. The surface morphologies of the ITO films prepared at 90 minutes (163 nm) sputtering time for specimens as-deposited and annealed at 300 °C in a vacuum are shown in Fig. 5. From the figures, the surface morphology of the annealed films seems to be very smooth. The value of surface roughness (Rms) was about 1.2 nm after annealing.

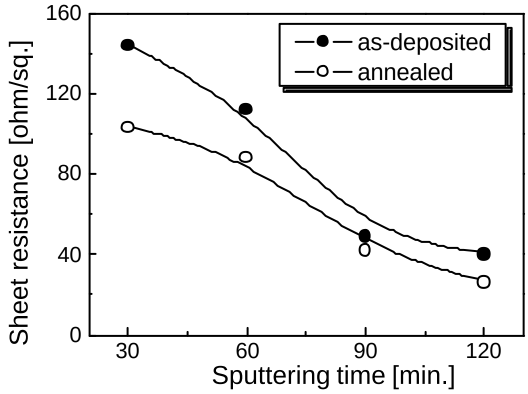
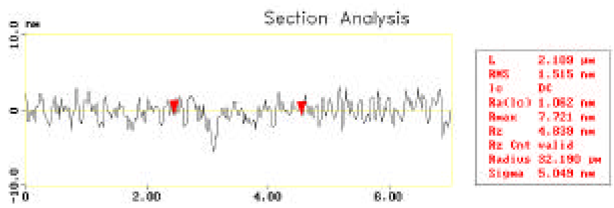
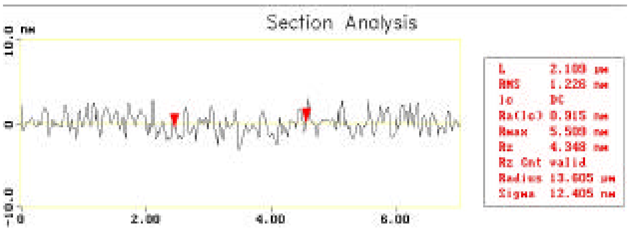


Fig. 4. The sheet resistance of ITO films as a function of the sputtering time, before and after annealing at 300 °C in a vacuum for about 10 minutes.



(a)



(b)

Fig. 5. The surface morphology of ITO films sputtered for 90 minutes (163 nm). The surface roughness of ITO film (a) as-deposited and (b) annealed at 300 °C for in a vacuum about 10 minutes.

4. Conclusions

In this study, ITO films were deposited on glass substrate using a LF (100 Hz) magnetron sputtering system and effect of thickness on the structural, electrical and optical properties of the films were investigated. According to the XRD analysis, the total crystallinity (I_{222}+I_{400}) of ITO films increased gradually with increasing of the film thickness (i.e. the sputtering time). The maximum transmittances in the visible region of all films exceed 80 % and the sheet resistance of the ITO films decreased with an increase of the film thickness. Additionally, the sheet resistance and the surface morphology of ITO films were also improved after annealing in a vacuum. As a result, we claim that the characteristics of ITO films grown by this way are strongly dependent on the film thickness.

5. Acknowledgements

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