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Effects of the cathode magnetic field strength on the properties of sputtered ultrathin ITO films

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Ultrathin indium tin oxide (ITO) films 20 nm thickness were manufactured by DC magnetron sputtering for touch sensor applications. Deposition was performed at room temperature on PET substrates, with a substrate-to-target distance of 50 mm, and a power and working pressure of 100 W and 0.28 Pa, respectively. This study focused on improving the electrical properties of ultrathin ITO films, which are typically poor. So, we controlled the initial growth process of the ITO films by optimizing the cathode magnetic field strength (CMFS) (550 G, 850 G, and 1450 G). The electrical properties of the thin films were optimal at the higher CMFS (1450 G) and the surface morphology and wetting properties also improved with strengthening CMFS. This is thought to be the result of a reduction in the damage incurred on the growing film as the plasma impedance and the high-energy particle (Ar^{O}, O^{-}) bombardment were reduced. Improved thin film properties can therefore be achieved by reducing high energetic particle (Ar^{O}, O^{-}) bombardment and increasing the number of sputter atoms with appropriate energy. In other words, the cathode-voltage (V_C) and cathode-current (I_C) during sputtering should be carefully adjusted.

Key words: Ultrathin ITO film, Touch panel, Cathode magnetic field strength, PET, Initial growth mechanism, DC magnetron sputtering.

Introduction

Indium tin oxide (ITO) is prepared by doping Sn with In_2O_3 and is a highly degenerated semiconductor with a wide band gap [1]. Because of its high electrical properties and optical transmittance in the visible range, ITO is a key component of a number of devices including flat panel displays, touch sensors, smart windows, flexible displays, and solar cells [1-3]. In particular, ultrathin ITO films have recently been the subject of much research due to the current tendency towards thinner, larger, and higher-resolution touch screen panels (TSPs). Generally, optical transmittance and sheet resistance of ITO thin film are trade-off relation and these are affected by initial growth process of thin film [4]. In other words, ultrathin ITO films with high optical transmittance have a high sheet resistance. As crystallinity affects the electrical properties of thin film, and their sheet resistance in particular. It can be improved by increasing the thickness of the films, but high crystallinity is rarely achieved in ultrathin ITO films [5, 6], Therefore, this is nonetheless crucial to improve the electrical properties of ultrathin ITO films. As crystallinity is affected by the processes-notably nucleation-occurring during the initial growth of the film, an understanding of the different parameters that affect these events is key. High electrical conductivity is difficult to achieve using existing sputtering processes, particularly at low temperatures. This is of relevance here since the polymer substrate used in this study is only suited to low temperature deposition (below 150 °C) because of its limited thermostability [7, 8]. A low-temperature deposition technique producing films with a low sheet resistance is therefore the objective of this study, wherein the effects of the cathode magnetic field strength (CMFS) are investigated on the initial growth of thin films. Adjusting the CMFS also allows the energy of the sputtered atoms to be controlled, which should improve the electrical properties of the ITO thin films. In other words, it is thought to be electrical properties of thin film can be improve as control sputter atom's momentum by establishing proper plasma impedance.

Furthermore, optimizing the CMFS is important in view of preserving the target. Ionization efficiency increases up to a certain level after which further increases only cause unnecessary damage to the target. For relatively weak CMFSs, the width of the etch track and the utilization of the target increase. But further strengthening of the CMFS provides more effective electron confinement and therefore increases the ionization efficiency. However, too strong a CMFS damages the target without any further increase in ionization efficiency. Controlling the CMFS is thereby also important in terms of preserving expensive targets [9]. In this research, therefore, the influence of the CMFS during initial film growth was investigated on the surface microstructure and electrical properties of sputtered thin films. Although the CMFS

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has been the subject of a number of previous studies, the influence of the CMFS on the initial growth of ultrathin ITO films is yet to be understood. Therefore, the results of this study have significance.

Experimental Details

To study the effects of the CMFS on the initial processes governing the growth of thin films, ultrathin (20 nm thick) ITO films were deposited on PET (polyethylene terephthalate) substrates at room temperature (RT) by DC magnetron sputtering (multi-sputtering system, Daeki Hi-tech Co.). High density ITO (In_2O_3 : 90 wt.%, SnO_2 : 10 wt.%) was used as a target and CMFSs of 550 G, 850 G, and 1450 G were applied, as measured from the center of the target. Sputtering was performed under 30 sccm flowing Ar and O_2 gas. The substrate-to-target distance was 50 mm. The DC power (power density) and the working pressure were 100 W (2.19 W/cm^3) and 0.28 Pa, respectively.

The thickness of the films was measured using a spectral reflectometer (ST-2000-DLXn, K-mac), their electrical properties were characterized through Hall-effect measurements (HMS-3000, ECOPIA), their optical transmittance by UV-visible spectroscopy (UV-vis, Shimadzu), and their surface roughness by atomic force microscopy (AFM, XE-100, PSIA Corp.). The contact angle of the thin films was measured using a contact angle meter (KRUSS Easy Drop) employing the sessile drop method. The deposition time was controlled to obtain 20 nm-thick films. The I-V characteristics of the cathode and the deposition rate were recorded as a function of the CMFS.

Results and Discussion

I-V characteristics and deposition rate

Figure 1a shows the I-V properties of the ITO films during sputtering process obtained at different CMFSs (550 G, 850 G, and 1450 G). The cathode current (I_C) and the cathode voltage (V_C) mean the Ar positive ion flux and the acceleration energy of the sputtering atoms. In strong CMFS, I_C and V_C increase and decrease, respectively, which, the number of trapped secondary electrons increases with the CMFS leading to an increased collision probability with Ar gas molecules, which in turn results in a higher plasma density [10]. These enhanced plasma density are significant for initial growth stages of the thin films because initial nucleation growth are affected by kinetic energy of sputtered atoms as well as high energetic particle such as negative oxygen ions (O^-) and reflected Ar neutral atom (Ar^O). This is thought to be relatively high values of V_C should lead to the sputtered atoms reaching the substrate with moderately high energies, thereby facilitating their migration in the film and enhancing its crystallinity. However, too high a V_C leads to the generation of high-energy particles such as negatively charged oxygen ions (O^-) and neutral reflected Ar atoms (Ar^O), which bombard on the film surface during the deposition [11]. In particular, the kinetic energy of the O is proportional to V_C and their collision with the substrate could damage the ITO thin films, leading eventually to degrade electrical properties. Therefore, these high energetic particles must be reduced that is, enhanced plasma density induced by strong CMFS can enhance the film crystallinity. As a result, high performance ultrathin ITO films can be fabricated by increasing CMFS.

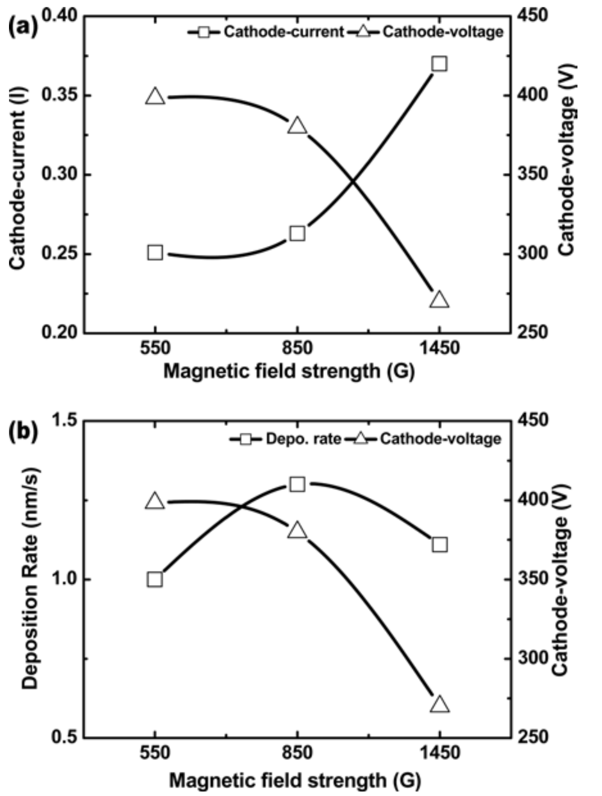

Fig. 1. (a) Cathode-voltage (VC) and cathode-current (IC) (b) Deposition rate and cathode-voltage (VC) of the ITO films as a function of different cathode magnetic field strength (CMFS) (550 G, 850 G, 1450 G).

Figure 1b shows the deposition rate of the films as a function of the cathode voltage at different CMFSs (550 G, 850 G, and 1450 G). The highest and lowest deposition rates are observed at 850 G and 550 G respectively. The deposition rate tends to increase with decreasing V_C up to 850 G, but decreases thereafter. The V_C at 550 G, 850 G, and 1450 G are 398 V, 380 V, and 270 V respectively, such that the difference in V_C at CMFSs of 550 G and 1450 G is 128 V. The low deposition rate at 550 G is due to the resputtering of atoms from the ITO films, which in turn is the result of high-energy particle bombardment [12]. However, when V_C is very low (at 1450 G), the kinetic energy of the positive Ar ions (Ar^+) and the sputtered atoms is insufficient to allow to migrate into the ITO films, thereby also leading to low deposition rates [13]. So, highest deposition rate was obtained for that of 850 G, due to enhanced plasma density while slightly reduced deposition rate revealed in that of 1450 G.

Electrical properties

Figure 2 shows electrical properties (resistivity, carrier density, and Hall mobility) in the ultrathin ITO films to observe the effect of different CMFSs (550 G, 850 G, and 1450 G) and it was deposited under a substrate-to-target distance of 50 mm, and a power and working pressure of 100 W and 0.28 Pa, respectively. The resistivity and sheet resistance (R_s) decrease sharply with increased CMFSs, while the carrier concentration and the Hall mobility increase. The R_s of the films prepared at 550 G and 850 G are 197 Ω/□ and 170 Ω/□, respectively. This variation in the resistivity of ultrathin ITO film could be explained by carrier density and Hall mobility. Interestingly, it was confirmed that both carrier density and Hall mobility were increased with increasing CMFS and it can be explained by improved crystalinity of ITO thin films. In the case of ITO films, electrical properties are mostly influenced by carrier density i.e. Sn^{4+} ions and oxygen vacancy (V_O), but Sn dopant is not activated in the amorphous structure of the ITO films, so V_O is dominant carrier, that is, the number of Sn^{4+} ions will be increasing as crystallized. Also, electrical properties of the ITO films are affected by the kinetic energy of sputtered atom during sputtering process. As, high energetic particles that bombard at the substrate can lead partial damage which can trap donors resultingly, it cause decline of doping efficiency. The lowest R_s, 127 Ω/□ was measured in the film prepared at 1450 G. This is thought to be slightly reduced the kinetic energy of the sputtering atoms but also reduced the number of damaging high-energy particles. This leads to improved growth during the initial stages, and thereby to a reduction in the R_s of the ITO films. On the other hands, the ITO ultrathin film deposited at 550 G has the highest resistivity and R_s. It is due to the damage incurred by the thin film from the increased number of high-energy particles (Ar^O, O^-) [14]. In summary therefore, the electrical properties of ultrathin ITO films can be improved by reducing the number of high-energy particles (Ar^O, O^-) while maintaining the kinetic energy of the sputtering atoms.

Figure 3 shows the R_s measured for ultrathin ITO films deposited at different O_2 ratios (in the gas flow) and CMFSs. It is examined to evidence variation of high energetic particles by different CMFS. The R_s is higher at 550 G than at 850 G or 1450 G. The R_s varies little up to an O_2 content of 0.2% but decreases thereafter. At 850 G, the R_s decreases slightly up to a concentration of 0.18% O_2 in the gas flow, but increases thereafter. In contrast, at 1450 G, the R_s increased dramatically for O_2 contents greater than 0.10%. That is, the R_s is minimal at 550 G, 850 G, 1450 G for O_2 contents of 0.22%, 0.18%, and 0.10%, respectively. The O_2 contents giving an optimal R_s decrease as the CMFS increases. That is, oxygen loss decreases by high energetic particles at strong CMFS. On the other hands, it is confirmed that carrier generation is dominated by V_O in the amorphous structure of ITO. So, optimal oxygen content is needed during deposition process to obtain improved electrical properties of ITO films, and to meet stoichiometry [15]. Meanwhile, the energy of the O^- ions is proportional to V_C so as the cathode voltage increases, they become more energetic than the reflected Ar atoms and deliver more momentum to the oxygen rather than to the In or Sn atoms. Consequently, the oxygen in the thin films is readily resputtered leading to the creation of a greater number of V_O carriers. The energy transfer function under O bombardment at the growing thin film surface is given by,

$$E = \frac{4M_O M_{O^-}}{(M_O + M_{O^-})} V_C \quad (1)$$

where V_C is the cathode-voltage (= initial energy of O^-),

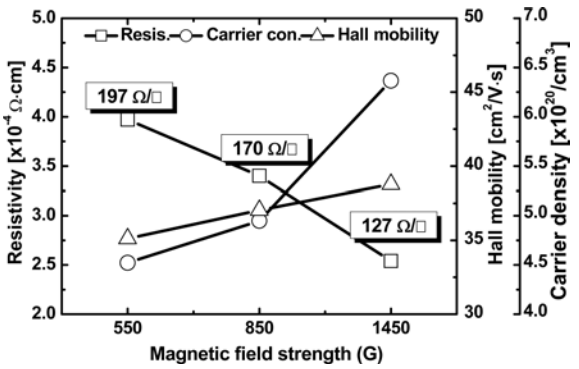

Fig. 2. Electrical properties (resistivity, sheet resistance, carrier concentration, Hall mobility) of the ITO films deposited 100 W, 50 mm, 0.28 Pa at RT by different cathode magnetic field strength (CMFS) (550 G, 850 G, 1450 G). The open square (□) indicates resistivity, open circle (○) and triangle (△) represent carrier density and Hall mobility, respectively.

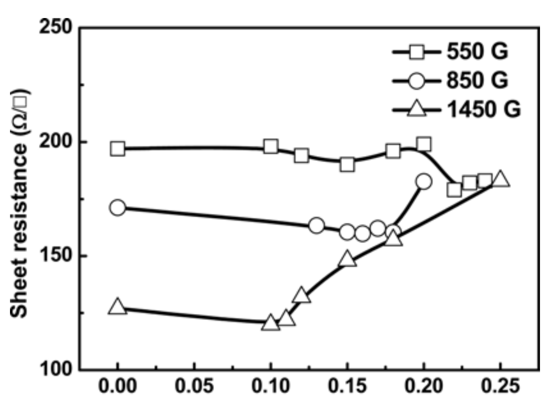

Fig. 3. O_2 flow ratio to have optimal sheet resistance of the ITO films deposited 100 W, 50 mm, 0.28 Pa at RT as a function of cathode magnetic field strength (CMFS) (550 G, 850 G, 1450 G).

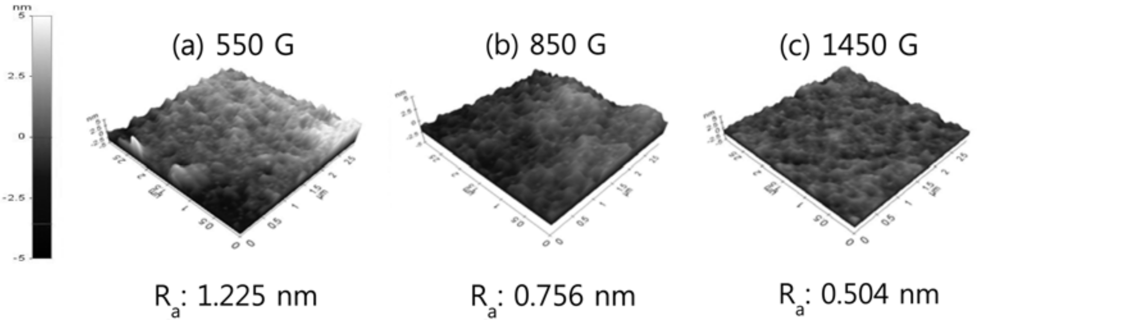

Fig. 4. AFM images of the ITO film deposited 100 W, 50 mm, 0.28 Pa at RT by different cathode magnetic field strength. (a) 550 G, (b) 850 G and (C) 1450 G).

M_o is a mass of an oxygen atom and M_{o-} is the a mass of a negative oxygen ion. As shown in equation 1, the transferred energy depends on the masses of the oxygen atoms and ions [10].

As shown in Figure 1a, V_C decreases with increasing CMFSs such that the number of high-energy O^- ions also decreases. To summarize therefore, the O_2 concentrations required to meet the stoichiometric conditions is smaller under stronger magnetic fields because fewer oxygen atoms are resputtered.

Morphology and contact angle

Figure 4 show AFM images of the ITO films prepared at CMFSs of 1450 G, 850 G, and 550 G. It is examined to observe initial growth stages of the ultrathin ITO films because it is similar Initial nucleation formation stages. The surface roughness (Ra) of the films is highest at the lowest CMFS, that is, 1.225 nm at 550 G compared with 0.504 nm at 1450 G.

As shown in Figure 1a, V_C increases as the CMFS is reduced, leading to increased bombardment of the films, as a result, partial damage on the substrate surface increase, result in nucleation formation density decrease, thereby to greater surface roughness. At a CMFS of 1450 G in contrast, the thin films incur less damage because of the lower V_C and the concomitant reduction in the number of high-energy particles [16, 17]. So, nucleation formation density is increased. Likewise, it is thought to be that surface roughness is affected by initial thin film growth stages, such as, surface damage and nucleation formation stages by sputter atom.

Figure 5 shows contact angles of ITO films deposited at different CMFS (550 G, 850 G, and 1450 G) to evidence surface roughness. The contact angle increases with the CMFS indicating a decrease in the wettability of the films.

Generally, low and high contact angles represent high and low surface energies, respectively. To stabilize a high-energy thin film surface, liquids must spread out over large areas. Furthermore, the surface energy decreases with Ra [18]. As shown Figure 4, the films produced at low CMFS have rough high-energy surfaces due to their bombardment by high-energy particles, and therefore, a low contact angle.

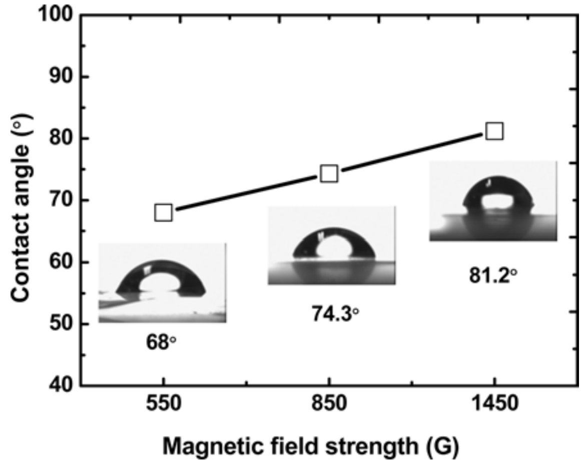

Fig. 5. Contact angle of the ITO films deposited 100 W, 50 mm, 0.28 Pa at RT by different cathode magnetic field strength (550 G, 850 G, 1450 G).

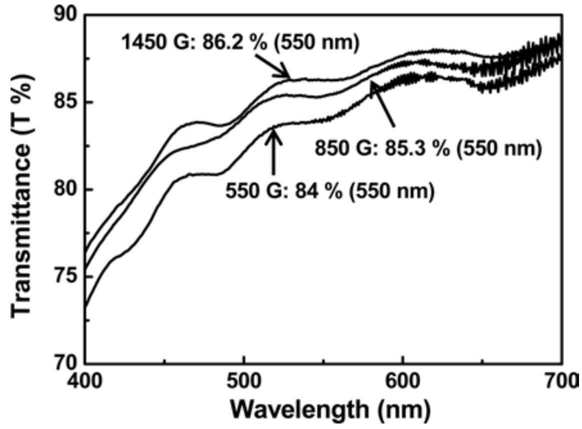

Fig. 6. Transmittance of the ITO films deposited 100 W, 50 mm, 0.28 Pa at RT as a function of cathode magnetic field strength (CMFS) (550 G, 850 G, 1450 G).

Optical properties

Figure 6 shows the optical transmittance of the films at different CMFSs (550 G, 850 G, and 1450 G). The optical transmittance increases with the CMFS, with the highest optical transmittance of about 86% obtained for the film prepared at 1450 G.

These results are attributed to the reduction of partial damage on the substrate of the ITO films by the stronger magnetic fields, which guarantee a composition closer to ideal stoichiometric conditions. It is thought to be that highly energetic particles disturb surface migration as increasing Ar^+. The optical transmittance in the visible range of a film depends on its microstructure and surface morphology [19]. Surface scattering increases with Ra such that the highest transmittance is obtained for the smoothest surfaces. This explains why the optical transmittance is optimal in the films prepared at 1450 G.

Conclusions

This study compared the deposition rate, I-V characteristics, electrical properties, and optimal transmittance of ultrathin ITO films prepared at different CMFSs (550 G, 850 G, and 1450 G). The surface roughness and contact angle of the films were also examined. The results obtained reflect the effect on the sputtering atoms of the CMFS. As the CMFS increases, the number of damaging high-energy particles (Ar^O, O^-) decreases while the number of sputtering atoms with an appropriate kinetic energy increases. The thin films produced at higher CMFSs are therefore smoother, more wettable, and have improved electrical and optical properties. In conclusion therefore, when manufacturing ultrathin ITO films, decreasing the number of high-energy particles and thereby reducing the damage incurred by the thin films is favorable to their initial growth, leading to the production of higher-quality thin films.

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