

Fabrication and characterization of indium tin oxide films

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ABSTRACT

Background: Transparent conducting oxide (TCO) films are of particular interest in the field of optoelectronics, due to the requirement for transparent electrodes in applications such as organic light-emitting diodes, solar cells and so on. The aim of this study was to obtain a better understanding of the effects of preparation temperature on indium tin oxide (ITO) films, to improve their performance for optoelectronic applications.

Methods: ITO films were deposited on glass substrate at different temperatures, using direct current (DC) magnetron sputtering. The influence of substrate temperature on the microstructure and electrical and optical properties was studied. The surface topography and microstructure of the films were analyzed by atomic force microscopy. The electrical resistivity and optical transmittance of the films were measured using the Hall effect measurement and spectrometer, respectively.

Results: The results showed that both the surface roughness and film thickness increased as the substrate temperature increased. Transmittance increased from 78% to 80% in the visible wavelength region, while resistivity decreased from 6.05×10^{-4} to 3.27×10^{-4} Ω -cm as the substrate temperature increased from 25°C to 275°C.

Conclusions: High-quality ITO films with low resistivity and high transmittance can be achieved by increasing the deposition temperature.

Keywords: Indium tin oxide, Optical transmittance, Resistivity, Substrate temperature

Introduction

Transparent conducting oxide (TCO) films are of particular interest in the field of optoelectronics, due to the requirement for transparent electrodes in applications such as organic light-emitting diodes (1), solar cells (2), thin-film transistor (TFT)-displays (3) and antireflection or antistatic coatings (4). Indium tin oxide (ITO) film, with low electrical resistivity, excellent optical transparency, high infrared reflectance and good chemical stability, is the most widely used transparent, conducting oxide film in optoelectronic devices. ITO films can be prepared by a range of different techniques, such as pulsed laser deposition (5), electron beam deposition (6), sputtering (7) and chemical vapor deposition (8). For optoelectronic applications, TCO film must be carefully fabricated to maximize optical transmittance in the visible region,

with minimum electrical resistivity (9). Optimization of the optical and electrical properties requires careful control of the fabrication processes. Sputtering with good reliability, high growth rate and better control of the film characteristics is one of the most often used methods for ITO film preparation (10). Tului et al (11) deposited ITO films onto glass substrates using magnetron sputtering under different atmospheres. They found that the presence of oxygen in the deposition chamber affected the growth direction of the crystal grains, which determined the conductivity of the ITO film. At high oxygen content, the films grew along the $\langle 111 \rangle$ direction, with high resistivity; at low oxygen content, the grains grew also along other orientations – e.g., the $\langle 100 \rangle$ direction – and the resistivity was decreased. Kavei and Mohammadi Gheidari (12) deposited ITO films on a glass substrate using the sputtering system with different sputtering gas pressures at room temperature. They reported that the optimum sputtering gas pressure of 27 mTorr provided a homogenous and most favorable rate of deposition, leading to good combined electrical conductivity and optical transparency. Meng et al (13) deposited ITO films onto polycarbonate substrate at room temperature by the ion beam-assisted deposition technique at different screen voltages. They found that the film prepared at low screen voltage showed an amorphous structure with a high optical transmittance, while the film prepared at high screen voltage showed a polycrystalline structure with a high electrical resistivity.

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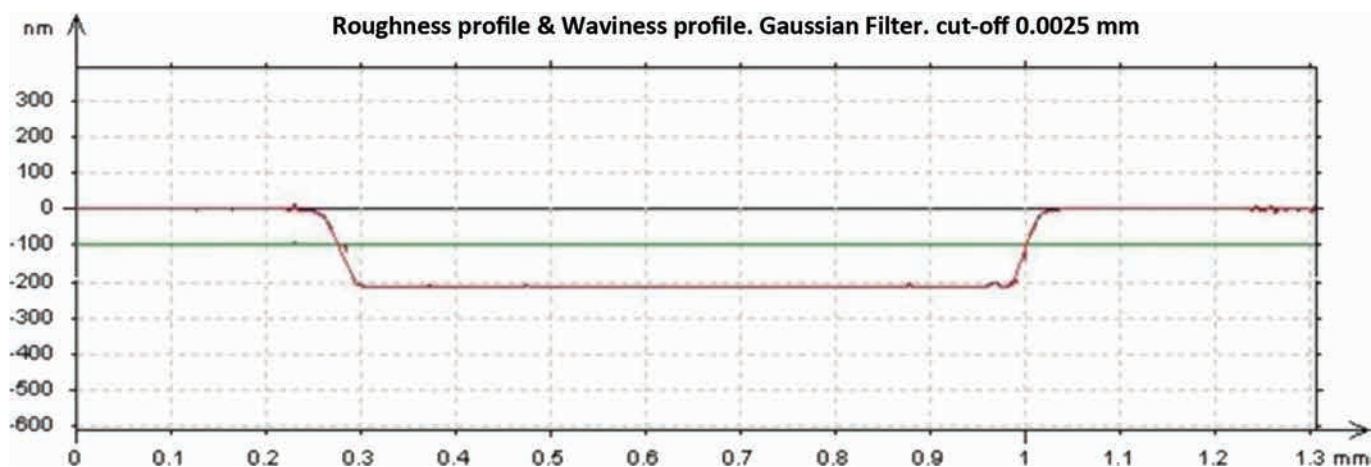


Fig. 1 - Surface profile of the indium tin oxide (ITO) film.

It is well known that the optical and electrical properties of sputtered ITO films are heavily dependent on the processing parameters, such as sputtering power, oxygen content and deposition pressure. In this present work, ITO films were grown onto glass substrate by direct current (DC) magnetron sputtering at different substrate temperatures. The effect of temperature on the morphology, electrical and optical properties of the ITO films was analyzed for different preparation conditions. The aim of this study was to obtain a better understanding of the temperature effect on the ITO films, to improve the performance for optoelectronic applications.

Methods

ITO films were deposited on 2-mm-thick glass substrate (Corning 2000) by DC magnetron sputtering system (ULVAC MB06-4703) at different substrate temperatures. A target (5-in. diameter, 3-mm thickness) that was In_2O_3 90% and SnO_2 10% in weight was used as sputtering source. The substrates were degreased in a dilute detergent solution, and rinsed in deionized water. Subsequently, they were subjected to ultrasonic cleaning in acetone solution, rinsed in an ultrasonic bath for 15 minutes and blow dried in hot air. After that, the substrate was placed in an oven to dry at a temperature of 50°C for 30 minutes, before it was introduced into the chamber. The normal distance between the substrate and target was about 10 cm. To obtain the required vacuum, a combination of rotary and turbo pumps was employed. The ultimate vacuum of 8×10^{-4} Pa was achieved after a pumping duration of 90 minutes. After that, highly pure argon (99.999%) was introduced into the chamber at the flow rate of 10 sccm, which acted as the sputtering gas. The working pressure in the chamber was kept at 2×10^{-1} Pa throughout the deposition. Prior to the deposition process, the substrate and target were pre-sputtered by Ar plasma for 20 minutes to remove any contaminants and to clean the surfaces. Following the same process as described above, 4 ITO films were grown on the glass substrate at 4 different substrate temperatures of 25°C, 100°C, 200°C and 275°C. The influence of substrate temperature on the surface topography, optical transmittance and electrical resistivity of the ITO film was studied. During all of

the depositions, the sputtering power and duration were kept constant at 100 W and 20 minutes, respectively. The thicknesses of all films were measured by a surface profiler (KLA P16; Tencor Co.). Before the deposition process, a lift-off tape was adhered to each of the 4 corners of the square substrate to create a small step on the substrate surface. After the deposition process, the tape was removed from the substrate, leading to an empty region in the substrate without the ITO film. The surface profile across the empty region was scanned using surface profiler. Figure 1 shows the surface profile with a step drop. The height of the step at the 4 corners of the substrate was measured, and the thickness of the film presented in this work is the mean value of these measurements. The thicknesses of the ITO films deposited at substrate temperatures of 25°C, 100°C, 200°C and 275°C were 203 nm, 209 nm, 221 nm and 238 nm, respectively. It appears that the thickness of ITO film increased as the substrate temperature increased.

Results and discussion

Topography of the ITO films

To investigate the possible correlation between the surface topography and optical and electrical properties of ITO films, atomic force microscopy (AFM; SPA 400; Seiko Instruments Inc.) was employed. Figure 2 shows the 3-dimensional AFM images of the ITO films deposited at different substrate temperatures. The roughness (measured in root mean square [RMS]) of the ITO films prepared at substrate temperatures of 25°C, 100°C, 200°C and 275°C were 0.331 nm, 0.393 nm, 1.008 nm and 1.440 nm, respectively. This shows that the surface roughness of ITO film increased as the substrate temperature increased. ITO films deposited at room temperature showed a very smooth surface with a RMS roughness of less than 0.4 nm. ITO films deposited at higher substrate temperatures were denser and rougher, with a maximum of 1.44 nm RMS roughness at 275°C. As the substrate temperature increased, more kinetic energy was introduced into the deposition atoms, leading to an increase of migration mobility, which in turn increased the surface roughness. The samples

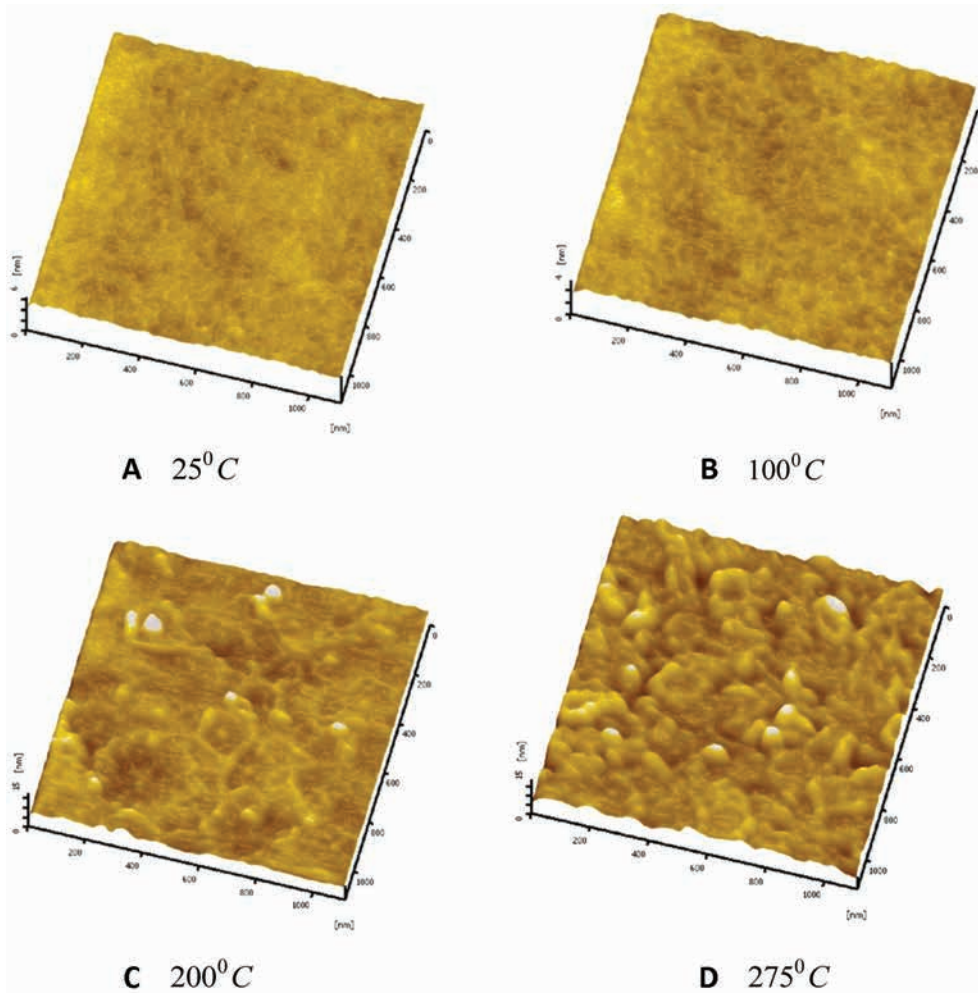


Fig. 2 - Surface topography of indium tin oxide (ITO) films deposited at different substrate temperatures.

grown at low temperatures had an amorphous nature, but the samples grown at higher temperatures showed good crystallinity. No apparent changes of the surface morphologies were observed when the films were deposited at a temperature lower than 200°C. This may be attributed to the uniform thermal treatment gained by the sample during the deposition process. When the substrate temperatures were higher than 200°C, the grains became distinctly larger by expanding into their neighbors and appearing as a number of stacked rods. The morphology of ITO films, as shown in Figure 2, exhibits uniformly distributed collar grains with fine crystallinity at higher substrate temperatures. A greater RMS value indicates an increase in mean crystallite size. An increase in the size of the crystallites with the increase of the substrate temperature was also evident from the AFM images. The surface topography of ITO film deposited at substrate temperature of 275°C exhibited a collar appearance, which reveals a characteristic high temperature growth feature in polycrystalline thin film.

Optical properties

Optical transmittance and reflectance of the ITO films were measured in the wavelength region between 300 nm and 1,200 nm, using a Lambda 750 from Perkin Elmer with

a double-beam spectrophotometer. The optical transparency of ITO film is heavily dependent on surface irregularity and defects such as pores. The optical measurements were evaluated as a function of the light wavelength for ITO films prepared at various substrate temperatures. The transmittance spectrum in the wavelength range of 300-1,200 nm is plotted in Figure 3. The sinusoidal behavior of the curves of transmittance vs. wavelength of light was due to interference phenomena between the wave fronts generated at the 2 interfaces (air and substrate) defined by the film. The average transmittance and reflectance of ITO film in the wavelength ranges of $\lambda_1 \sim \lambda_2$ can be calculated as follows:

$$\bar{T} = \frac{\int_{\lambda_1}^{\lambda_2} T d\lambda}{\lambda_2 - \lambda_1} \tag{Eq. [1]}$$

$$\bar{R} = \frac{\int_{\lambda_1}^{\lambda_2} R d\lambda}{\lambda_2 - \lambda_1} \tag{Eq. [2]}$$



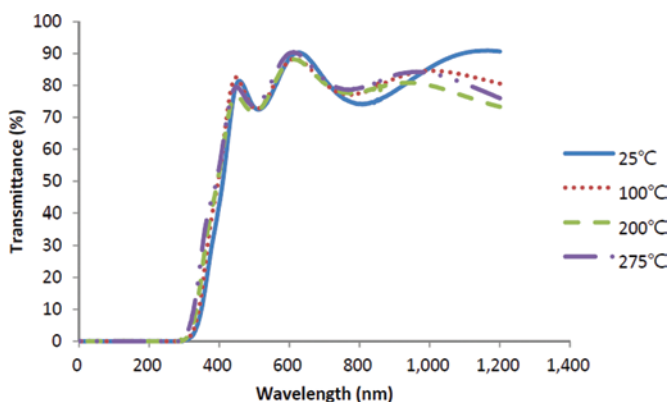


Fig. 3 - Optical transmittance spectra of indium tin oxide (ITO) films prepared at various substrate temperatures.

TABLE I - Average optical transmittances in the wavelength ranges of 300-1,200 nm and 400-800 nm for indium tin oxide (ITO) films prepared at various substrate temperatures

Wavelength range	Substrate temperature			
	25°C	100°C	200°C	275°C
300-1,200 nm	72.20%	73.88%	74.06%	74.84%
400-800 nm	78.45%	78.39%	79.25%	80.28%

The average transmittances of ITO films deposited at different substrate temperatures are listed in Table I. It shows that the average transmittance of ITO film increased from 72.2% to 74.8% in the wavelength region of 300-1,200 nm, and increased from 78.5% to 80.3% in the visible wavelength region of 400-800 nm, as the substrate temperature increased from 25°C to 275°C. The reflectance spectra of ITO film are plotted in Figure 4. Table II lists the average reflectance of ITO films. It shows that the average reflectance of ITO film decreased from 17.6% to 13.9% in the wavelength region of 300-1,200 nm, and decreased from 19.8% to 16.6% in the visible wavelength region of 400-800 nm, as substrate temperature increased from 25°C to 275°C.

Bandgap energy

The optical bandgap energy of ITO film is calculated by applying the Tauc model and the Davis and Mott model (14).

$$\alpha h\nu = A(h\nu - E_g)^n \tag{Eq. [3]}$$

Where α , $h\nu$ and E_g denote the absorption coefficient, photon energy and bandgap energy, respectively. A is a constant independent on the photon energy, and n is a constant depending on the nature of electronic transition. In this work, the value of n is 0.5 because the ITO film is a direct transition material. Substituting $n = 0.5$ into Equation (3), yields

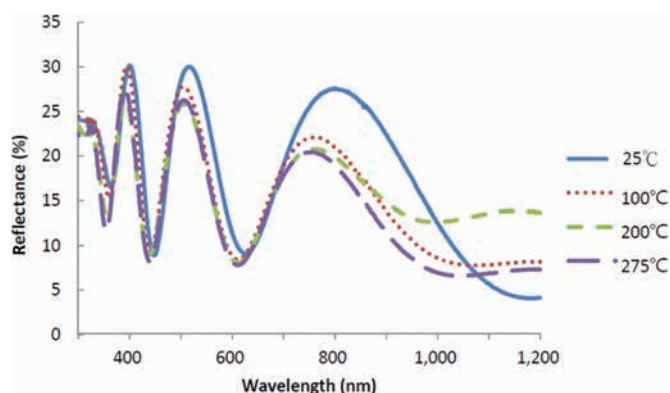


Fig. 4 - Optical reflectance spectra of indium tin oxide (ITO) films prepared at various substrate temperatures.

TABLE II - Average optical reflectances in the wavelength ranges of 300-1,200 nm and 400-800 nm for indium tin oxide (ITO) films prepared at various substrate temperatures

Wavelength range	Substrate temperature			
	25°C	100°C	200°C	275°C
300-1,200 nm	17.56%	15.43%	16.11%	13.88%
400-800 nm	19.81%	17.94%	16.73%	16.60%

$$(\alpha h\nu)^2 = A^2(h\nu - E_g) \tag{Eq. [4]}$$

The absorption coefficient α is determined from the following relation (15):

$$\alpha = \frac{1}{d} \ln\left(\frac{1}{T}\right) \tag{Eq. [5]}$$

Where T and d are the optical transmittance and film thickness, respectively.

The bandgap energy of the films can be determined by extrapolation of the straight-line portion of the plot of $(\alpha h\nu)^2$ versus the photon energy $h\nu$ as shown in Figure 5. The photon energy at the point where $(\alpha h\nu)^2 = 0$ is denoted as the bandgap energy E_g . Figure 5 shows the plot for ITO film deposited at substrate temperature of 200°C. The bandgap energies of ITO films grown at substrate temperatures of 25°C, 100°C, 200°C and 275°C were 3.527, 3.625, 3.754 and 3.8 eV, respectively. The bandgap energy of ITO films varied along with the substrate temperature. It increased from 3.527 to 3.8 eV as the substrate temperature increased from 25°C to 275°C. The bandgap can be changed by the alterations in the oxygen sites. The increase in the bandgap may be associated with decreases in oxygen content in the film that might have been caused by the higher substrate temperature. As the substrate temperature increased, more oxygen was evaporated in a process that could cause a decrease in the oxygen content



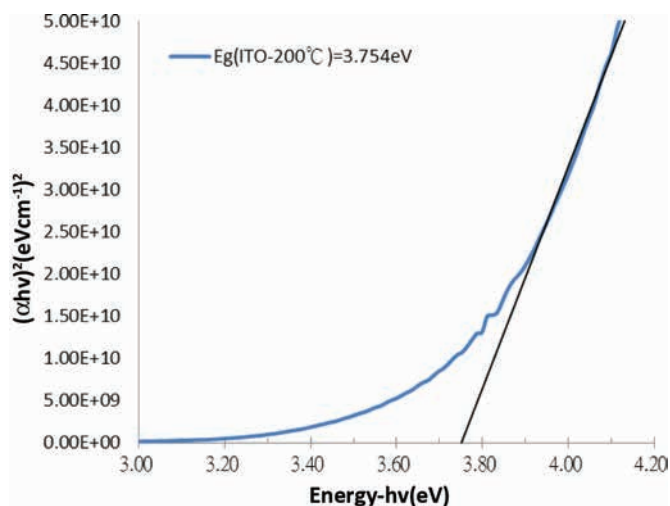


Fig. 5 - Plot of $(\alpha hv)^2$ versus $h\nu$ for indium tin oxide (ITO) film prepared at substrate temperature of 200°C.

in the films. It can be observed that the bandgap energy increases as the substrate temperature increases. The wide bandgap leads the shift of the absorption edge to a shorter wavelength as shown in Figure 3 due to the Burstein-Moss effect. According to the Burstein-Moss effect, the increase of the Fermi level in the conduction band leads to the bandgap energy broadening with increasing carrier concentration. The blue shift of the absorption edge is attributed to the increasing crystallinity quality of the ITO films deposited at higher substrate temperature.

Electrical properties

The electrical properties such as resistivity, carrier concentration and mobility were determined from Hall effect measurements at room temperature using an Ecopia HMS-300 system based on the van-der-Pauw method. The measurements of electrical resistivity for each sample were repeated several times. The resistivities of ITO films deposited at substrate temperature of 25°C, 100°C, 200°C and 275°C were 6.054×10^{-4} , 5.065×10^{-4} , 4.148×10^{-4} and 3.274×10^{-4} Ω-cm, respectively. The carrier concentration increased from 3.294×10^{20} to 8.158×10^{20} as the temperature increased from 25°C to 275°C as shown in Figure 6. The negatively charged oxygen species formed depletion regions near the grain boundary surfaces which acted as trapping sites and formed the potential barriers to the carriers. The depletion regions in ITO films deposited at higher substrate temperature can be removed from the grain boundary surface results in an increase of the carrier concentration. The electrical properties have good correlation with their optical properties related to bandgap energy. The mobility of ITO film varying with substrate temperature is plotted in Figure 7. The electrical properties including the resistivity, carrier concentration and mobility of ITO films prepared at temperatures of 25°C, 100°C, 200°C and 275°C are listed in Table III. The resistivity of ITO films decreased with increasing substrate temperature, as shown in Figure 8. The effect of the substrate temperature on the

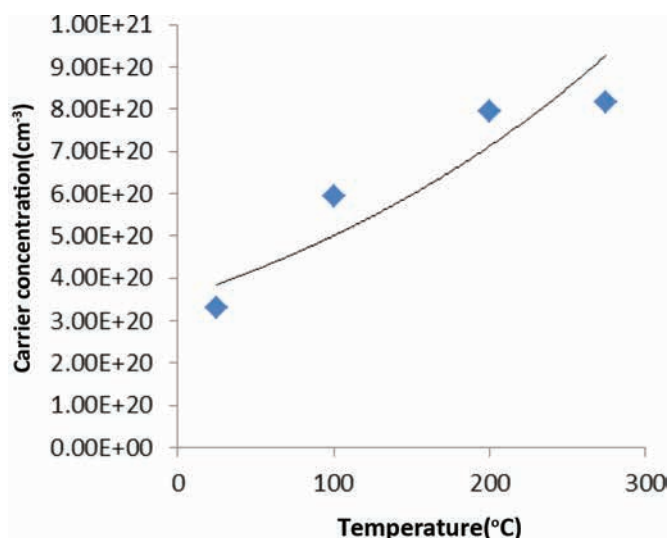


Fig. 6 - Carrier concentration of indium tin oxide (ITO) film varied with substrate temperatures.

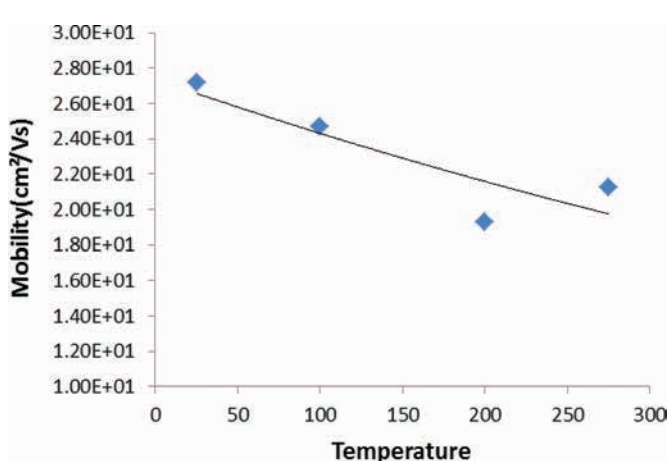


Fig. 7 - Mobility of indium tin oxide (ITO) film varied with substrate temperatures.

TABLE III - Electrical properties of indium tin oxide (ITO) films prepared at various substrate temperatures

Electrical properties	Substrate temperature			
	25°C	100°C	200°C	275°C
Resistivity (10^{-4} Ω-cm)	6.054	5.065	4.148	3.274
Carrier concentration (10^{20} cm ⁻³)	3.294	5.946	7.957	8.158
Mobility (cm ² /Vs)	27.16	24.71	19.31	21.23

resistivity was mainly based on an enhancement of the carrier concentration.

Conclusions

ITO films were deposited on glass substrates by radio frequency magnetron sputtering at different substrate



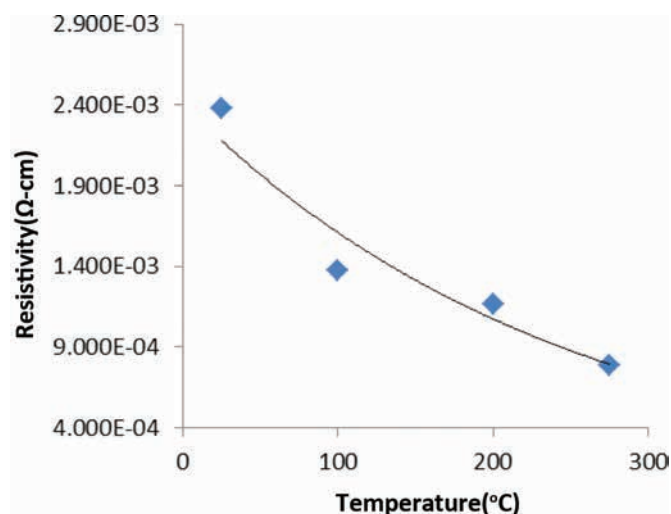


Fig. 8 - Resistivity of indium tin oxide (ITO) film varied with substrate temperatures.

temperatures varying from 25°C to 275°C. This study investigated the influence of the substrate temperature on the microstructure and optical and electrical properties of ITO films. The microstructure was characterized by AFM. It was observed that the crystallinity was improved by increasing the substrate temperature. The optical transmittance increased as the substrate temperature increased. Experimental results showed that average optical transmittance of 80% in the visible region was achieved for ITO film prepared at substrate temperature of 275°C. The electrical resistivity decreased, and the carrier concentration increased, as the substrate temperature increased. It was found that the improvement of electrical conductivity along with the optical transmittance enhancement of ITO film were attributed to higher carrier concentration, film crystallite improvement and grain size enlargement at high substrate temperatures.

Disclosures

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Conflict of interest: None of the authors has any financial interest related to this study to disclose.

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