# The effect of electro-annealing on the electrical properties of ITO film on colorless polyimide substrate

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#### **Abstract**

The effect of different annealing methods on the sheet resistance of indium tin oxide (ITO) on polyimide (PI) substrate has been investigated. As electro-annealing induced the predominant growth of crystallites of ITO thin films along (4 0 0) plane, the sheet resistance of ITO films that were electro-annealed for 2 mA at 180  $^{\circ}$ C considerably decreased from 50 to 28  $\Omega$ /cm².

#### 1. Introduction

Transparent polymer substrates offer substantial advantages in terms of being able to develop displays that are thinner, lighter, more robust, deformable and can be rolled up when not in use. The ITO thin films that were deposited on polymer substrates, however, showed a high level of electrical resistance because substrate heating is limited due to its low thermal stability. As a result, polymer substrates with high glass transition temperature have been mainly investigated for their resistance to higher temperature during deposition [1,2].

Also, annealing experiments of ITO thin films on polymer substrates have been implemented and several researches confirmed that annealing temperature affects the microstructure, electrical and optical properties of ITO thin films [3,4].

A detailed investigation of the electrical and optical properties of ITO thin films on polymer substrates after electro-annealing, however, has not been conducted. In this paper, we have investigated the electrical and optical properties of ITO thin films on polyimide substrates as a function of electro-annealing conditions. The effect of electro-annealing on the microstructures and sheet resistance of the ITO thin films was investigated.

# 2. Experimental

A commercial colorless polyimide (PI, Mitsubishi Gas Chemical, Neopulim-L), that has thickness of  $200\pm10~\mu m$ , surface roughness of 1 nm and  $T_g$  of  $300~^{\circ}{\rm C}$  was used as a substrate for the deposition of ITO films. ITO thin films were prepared by RF magnetron sputtering of an oxide ceramic target (In<sub>2</sub>O<sub>3</sub>/SnO<sub>2</sub> =  $90/10~{\rm wt\%}$ ) at room temperature. The temperature of PI substrate rose up to  $60~^{\circ}{\rm C}$  during sputtering. The distance from target to substrate was  $100~{\rm mm}$ . A base pressure below  $5.0\times10^{-6}$  Torr was maintained prior to the deposition. High purity Ar was used as the sputtering gas and the process pressure was kept at  $2.1\times10^{-3}$  Torr.

ITO films that had thickness of 280 nm were obtained at 75 W for 20 min at room temperature and these factors were used for all experiments. The electro-annealing of the ITO thin films was conducted at a constant current flow (2 mA), which was heated from 100 to 180 °C for 60 min and cooled in ambient to room temperature conditions in air. Thermal annealing was carried out in the same manner was that of electro-annealing.

The microstructure of the ITO thin films was investigated by X-ray diffractometer (Phillips X'Pert APD) with CuKα (1.5418 Å) radiation. Sheet resistance was measured using a four-point probe (Mitsubishi Chemical, MCP-T610). The optical transmittance was observed using a UV–vis spectrometer (Jasco V-650). Field emission scanning electron microscope (HITACHI, S-4300) was used to observe the surface morphology and cross-sectional views of the ITO thin films.

### 3. Results and discussion

ITO-coated PI films were prepared at room temperature and annealed by electro- and thermal methods. The effect of annealing on the sheet resistance, microstructure and transmittance of the ITO-coated PI films were investigated. The sheet resistance of the annealed ITO thin films, as a function of annealing temperature, is plotted in Fig. 1.

The sheet resistance of the electro-annealed ITO thin films was slightly lower than that of thermal annealed ones at the same temperature. The sheet resistance of the annealed ITO thin films sharply decreased from 50 to  $28~\Omega/\text{cm}^2$  with increasing the annealing temperature. Because of the ITO film was crystallized uniformly and the crystalline ITO was enlarged at high annealing temperature. As a result, the sheet resistance or ITO films decreased in correlation with the growth of crystalline.

Through the X-ray diffraction patterns of ITO thin films which were both annealed and non-annealed were analyzed the intensity ratio of the (4 0 0) to (2 2 2) planes,  $I_{4\,0\,0}/I_{2\,2\,2}$ , as a function of the annealing temperature, is shown in Table 1. The grain size was derived from the XRD spectra following Debye–Scheerer's method [5], and the results are summarized in Table 1.

The intensity of (2 2 2) plane diffraction peak was higher than that for other planes in all films. The electroand thermal annealed ITO thin films are made of polycrystalline with a preferential orientation along (2 2 2) plane. Also, annealed ITO thin films were oriented along (4 0 0) plane. The sheet resistance of

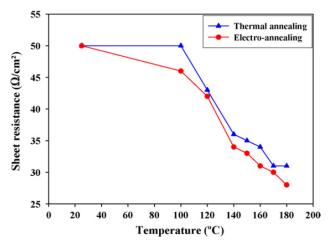


Fig. 1. Changes in sheet resistance after annealing at various temperatures (deposition condition: 75 W; RT: 20 min).

TABLE 1. Effect of annealing the sheet resistance and microstructure of the ITO crystallites

and microstructure of the ITO crystallites				
	Sheet resistance $(\Omega / cm^2)$	$\frac{I_{400}}{I_{222}}$	Grain size <sup>a</sup> (Å)	Grain size <sup>b</sup> (Å)
Non-annealing	50	1.21	180	251
Thermal annealing (150 °C)	35	0.51	199	267
Electroannealing(150 $^{\circ}$ C)	33	0.84	195	269
Thermal annealing (180 $^{\circ}$ C)	32	0.58	203	268
Electro-annealing (180°C)	28	0.66	198	270

<sup>&</sup>lt;sup>a</sup> Calculated from the (2 2 2)

the electro-annealed ITO thin films was slightly lower than that of the thermal annealed ones at the same temperature, because the electro-annealed ITO thin films were more oriented along (4 0 0) plane.

The intensity ratio of the  $(4\ 0\ 0)$  to  $(2\ 2\ 2)$  planes  $(I_{4\ 0}$  $_0/I_{2\,2\,2}$ ) was used to evaluate the annealing effect on the crystalline texture of ITO films. The values of  $I_{400}$ /  $I_{2\,2\,2}$  decreased after electro- and thermal annealing. It is well documented that (2 2 2) plane of the crystalline ITO was predominantly enlarged due to thermal annealing and the sheet resistance of the ITO films depends upon the orientation of (4 0 0) plane rather than that of (2 2 2) plane. The  $I_{400}/I_{222}$  of the thermal annealed ITO films was lower than that of the electro-annealed ITO films. The thermal annealed ITO films were predominantly oriented along (2 2 2) plane. Whereas the crystallites of the electro-annealed ITO films grew along (4 0 0) plane, the sheet resistance of the electro-annealed ITO films was lower than that of the thermal annealed ITO films.

The grain size of the crystalline ITO was 20 and 27 nm which were calculated from (2 2 2) and (4 0 0) plane, respectively. The grain size of the electroannealed ITO thin films is bigger than that of the thermal annealed one in (4 0 0) plane, whereas the grain size of the thermal annealed ITO thin films is bigger than that of the electro-annealed ones in (2 2 2) plane. These results correspond with the growth of crystallites and the intensity ratio of (4 0 0) to (2 2 2) planes of the ITO films by electro- and thermal annealing. The mean free path, which is a path whereby electrons can move freely in metal, is

<sup>&</sup>lt;sup>b</sup> Calculated from the (4 0 0)

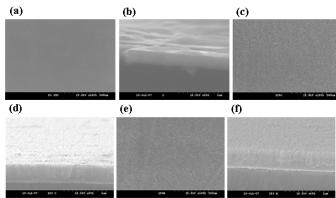


Fig. 2. SEM micrographs of the ITO films on PI before and after annealing: (a) surface morphology and (b) cross-section before annealing; (c) surface morphology and (d) cross-section after thermal annealing (at 150  $^{\circ}$ C); (e) surface morphology and (f) cross-section after electro-annealing (at 150  $^{\circ}$ C).

lengthened by a decrease in grain boundaries and by an induction in the decrease of sheet resistance. The increase in the grain size brings about a decrease in the grain boundaries. Sheet resistance decreases with an increase in grain size [6–8]. According to these results, electro-annealing has led to the growth of the crystallites along (4 0 0) plane and enlarged the grain size of ITO films.

In observing the changing crystalline in ITO thin films, surface morphology and cross-sections of the ITO thin films were investigated with SEM images are shown in Figure 2. The surface morphology and cross-sectional microstructure of non-annealed ITO film manifested a very smooth surface and could not be seen crystallites, however, the surface morphology of the electro- and thermal annealed ITO film (Figure 2(c) and (e)) showed microcrystalline points such as grain aggregates. The cross-section of the electro-annealed ITO film clearly showed a polycrystalline phase and grains with a columnar structure.

## 4. Summary

ITO thin films were deposited onto PI substrate by RF magnetron sputtering at 75 W, room temperature for 20 min. The effect of electro- and thermal

annealing on the microstructural, optical and electrical properties of ITO thin films has been investigated. Sheet resistance considerably decreased by annealing over 150  $^{\circ}$ C. At 180  $^{\circ}$ C, the sheet resistance of the electro-annealed ITO film showed a considerable reduction from 50 to 28 V/cm<sup>2</sup>. The electro-annealed ITO thin films increased predominant growth of crystallites of ITO films along (4 0 0) plane, on the other hand, the crystallites of the ITO films grew along (2 2 2) plane by thermal annealing. The transmittance of ITO thin films was slightly improved by electroannealing because of the enlarged grain size of the electroannealed ITO films. The electroannealing method is quite effective in reducing the sheet resistance of ITO thin films that are coated onto PI substrate. Those of the ITO thin films that were coated onto PI films can be expected to be applied to the substrates of flexible displays.

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