Characterization of ITO thin films on PET substrates prepared by gas-timing RF magnetron sputtering

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Indium tin oxide (ITO) thin films were deposited on polyethylene terephthalate (PET) substrates at room temperature by RF magnetron sputtering with a new technique, called gas-timing. The influence of RF power on properties of ITO thin films was investigated. From the x-ray diffraction measurement of the ITO thin films grown by this technique, cubic nano-crystalline structure with predominant (222) and (400) orientation was observed. It was found that the increasing of RF power yielded to decrease in the sheet resistance of ITO thin films. The sheet resistance of ITO thin films deposited by RF power of 40 watts had the lowest value of 8 Ω/□. By the gas-timing technique, the ITO thin films on PET substrates were achieved with high transmittance in visible region of 90%. Therefore, the gas-timing RF magnetron sputtering is a promising technique to achieve the ITO thin film on PET substrate with low resistivity and high transmittance in visible region without substrate heating and post-deposition annealing. [DOI: 10.1380/ejssnt.2005.272]

Keywords: Indium tin oxide; RF magnetron sputtering; Gas-timing; Polyethylene terephthalate (PET)

I. INTRODUCTION

Tin doped Indium oxide (ITO) is an n-type, highly degenerated, wide band gap semiconductor with relatively low resistivity and high transmittance in visible range region [1]. Due to these properties, it has been widely used as transparent electrodes in various displays, including liquid-crystal display (LCD), electro luminescent display (ELD), and organic light-emitting diode (OLED) [1]. It has been reported that RF magnetron sputtering yields ITO thin films with low resistivity and good reproducibility. For the crystallization of ITO thin films, the heating process at an elevated temperature during the film deposition or an addition post-annealing treatment at around 150°C is required [2]. The substrate temperature is an important parameter for several applications, particularly when organic polymer substrates are used [3, 4]. By this technique, we have succeeded in growing AlN at room temperature [5]. In this report, the deposition and characterizations of ITO thin films on PET substrate by RF magnetron sputtering with gas-timing technique are investigated.

II. EXPERIMENT

The ITO thin films were deposited on polyethylene terephthalate (PET) substrates using the rf magnetron sputtering system. A ceramic In$_2$O$_3$:SnO$_2$ target (90:10 wt.%, 99.99% purity) from Kurt J. Lesker was used. PET substrates were cleaned by alcohol process. The sputtering system was pump down to $1 \times 10^{-6}$ mbar using diffusion pump. Working pressure consisted mainly with high-purity Ar (99.99%) gas was $3.2 \times 10^{-3}$ mbar. The distance between target and the substrate was 5 cm, and the RF power varied from 10 W to 40 W, whereas the thickness of ITO thin films was 200 nm. The deposition process was carried out at room temperature, i.e. the substrate was not heat during or after the film deposition. ITO thin films were also deposited intermittently. The pure Ar gas was fed to the system for sputtered shows in Fig. 1.

![Gas-timing technique for ITO thin films growth. The pure Ar gas was fed to the system for 50 seconds (Ar gas turned on) and stop it for 2 seconds (Ar gas turned off).](image-url)

TABLE 1: Sputtering conditions used for deposition of ITO thin films.

| Condition            | Value               |
|----------------------|---------------------|
| Base pressure        | $1.0 \times 10^{-6}$ mbar |
| RF power             | 10, 20, 30, 40 Watt  |
| Substrate-target distance | 50 mm               |
| Substrate temperature | Room temperature    |
| Sputtering pressure  | $3.2 \times 10^{-3}$ mbar, Ar gas activated |
| Ar flow rate         | 10 sccm, fixed      |
| Ar Gas-timing (on:off) | 50:2 second         |

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pure Ar gas was fed to the system for 50 seconds (Ar gas turned on) and stop fed 10 seconds (Ar gas turned off) until the thickness of thin films was 200 nm. The turned on/off of Ar gas was controlled by computer. The 200 nm ITO thin films were grown on the PET substrates with several of sputtering conditions shown in Table 1. The crystalline structure was investigate by a X-ray diffraction (XRD) spectrometer (D8 Advance, Bruker) using Cu-K\textalpha radiation. The optical transmittance of the films was measured with a single beam spectrophotometer; scanned in the UV-Visible region (300$\sim$1000 nm). The optical transmission spectra of ITO thin films with various RF power show that the films are highly transparent invisible region, as shown in Fig. 4. The average transmission was found over than 90%

The optical bandgap of the films were determined by extrapolating the linear portion of the $h\nu$ versus $(\alpha h\nu)^{-2}$ curve to $(\alpha h\nu) = 0$ (inset of Fig. 5.). The absorption coefficient ($\alpha$) was determined from the relation, $I = I_0 \exp(-\alpha t)$ where $t$ is the thickness of the sample, $I$ the transmitted intensity at a particular wavelength and $I_0$ the maximum transmitted intensity which is taken to be 100%. This relation give $\alpha = (1/t) \ln(I_0/I)$ [6].

The bandgap of ITO films increase with increasing RF power, as shown in Fig. 5. The increase in bandgap may due to an increase in carrier concentration with increasing RF power as a result of which the absorption edge shift toward the near UV range [7]. The increasing bandgap with carrier concentration can be explain on the basis of Burstein-Moss effect. Assuming that the conduction band and valence band are parabolic in nature and that Burstein-Moss shift is the predominant effect, we can write $E_g = E_{g0} + \Delta E_{g}^{B-M}$ where $E_{g0}$ is the intrinsic bandgap and $\Delta E_{g}^{B-M}$ the Burstein-Moss shift due to the filling of low lying levels in the conduction band [8]. An expression for Burstein-Moss shift is given by $\Delta E_{g}^{B-M} = (h^2/8\pi^2m^*_e)(3\pi^2n)^{2/3}$ where $n$ is the carrier
FIG. 5: Variation of bandgap of the ITO thin films on PET substrate deposited with the gas-timing technique as a function of RF power. Inset shows a typical plot of $h\nu$ vs. $(a\nu)^2$ for the ITO film deposited with the gas-timing technique.

FIG. 6: Variation of sheet resistance of the ITO thin films on PET substrate deposited with the gas-timing technique as a function of RF power.

concentration and $m^*_v$ the reduced effective mass of the carriers. From this expression it is clear that Burstein-Moss shift is directly proportional to $n^{2/3}$. However, at very high carrier concentrations it is seen that there is bandgap narrowing due to electron-electron scattering and electron-impurity scattering [6].

The increase in carrier concentration may be due to an increase in the diffusion of Sn atoms from interstitial locations and grain boundaries into the In cation sites. Since the Sn atom has a valency of 4 and In is trivalent, the Sn atoms act as donors in ITO films [6]. Hence the increase in Sn diffusion with RF power results in higher electron concentration. In the addition, The gas-timing technique will be increased oxygen vacancy due to decreasing oxygen atoms that distributed from the center of target then it was pumped down when Ar gas turned off. The oxygen vacancy can donate two free electrons for conduction [6]. Hence the increase of oxygen vacancy with gas-timing technique result in higher electron concentration.

The electrical properties of the ITO thin film depend on the film composition and deposition parameters such as RF power, sputtering pressure, etc [9]. In this study, we found that the sheet resistance decrease with increasing in RF power. The minimum of sheet resistance was about 8 $\Omega/\square$ at RF power of 40 W shown in Fig. 6. This result was supported by the increase of carrier concentration when increased RF power that yields to reduction of sheet resistance [9].

IV. CONCLUSIONS

The 200 nm ITO thin film were prepared on PET substrates by RF magnetron sputtering with gas-timing technique at sputtering pressure of $3.2 \times 10^{-3}$ mbar. The crystallization, surface morphology, sheet resistance and transmittance of all samples were investigated by x-ray diffraction spectrometer (XRD), scanning electron microscope (SEM), four-point probe and UV-VIS spectrophotometer, respectively. The XRD patterns of all deposited ITO thin films showed orientation of cubic crystalline structure in (222) and (400) planes. The SEM images revealed that all ITO samples have the grain size smaller than 100 nm. The sheet resistance of ITO thin films is reduced from 30 to 8 $\Omega/\square$ as the RF power of sputtering increases from 10 W to 40 W. The ITO thin films on PET substrates have the transmittance of about 90% in visible region. Therefore, the ITO thin film on PET substrate with low resistivity and high transmittance in visible region has been achieved without substrate heating and post-deposition annealing by the novel gas-timing RF magnetron sputtering.

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