

Development of Textured ITO Optical Windows for Photovoltaic Applications

^{1*} Dr. SRIKANT KUMAR DASH ² Mrs. MITALI MOHAPATRA

^{1*} Asst. Professor, Dept. Of Electrical Engineering, NIT BBSR,

Asst. Professor DEPT. of Electrical Engineering, NIT BBSR,

srikantakumardash@thenalanda.com, mitalimohapatra@thenalanda.com

ITO thin films were deposited on glass substrates using rf magnetron sputtering. Deposition parameters such as the rf power and the oxygen dilution to the plasma are investigated. Films deposited with an increase in rf power were resulting in the film preferential orientation along (100) plane. Oxygen dilution to the plasma at low rf power of 30 W was found to result in a change in preferential orientation from (100) plane to (111) plane. SEM analysis revealed a spotty surface morphology for the (100) films and a granular morphology for the (111) films. The highest transmittance was found for the oxygen diluted (111) film. The highest optical bandgap of 4.33 eV was observed for the pure Ar deposited film. Details are discussed.

1. Introduction

Transparent conducting oxide (TCO) thin films have been widely investigated towards the application to solar cell fabrications due to their displayed characteristics of high optical transparency and better electrical conductivity along with environmental stability [1–3]. Basically, the optical and the electrical properties of TCOs depend solemnly on their oxidation conditions. Complete oxidation was found to promote the transparency in the UV-visible range of electromagnetic spectrum, and, in contrary, the incomplete oxidation was found to promote the electronic conductivity [4]. Hence, a careful adjustment between the oxidation and the oxygen vacancy doping paves the way for the attainment of optimum values of optical transparency with optimum electrical conductivity [5].

In this context, indium tin oxide (ITO), a wide bandgap (>3.6 eV) degenerate semiconducting material, shows the highest value of optical transparency greater than 90%, better polycrystalline structures with either (222) or (400) preferred orientation, and the lowest electrical resistivity in the order of 10^{-5} Ω -cm [4, 5]. Specially, the highest possible carrier doping density in the order of 10^{21} cm^{-3} and the associated localization of Fermi level above the conduction band level boost the very low resistivity and high transmittance in the visible region [6, 7]. There are various deposition techniques such as DC and rf magnetron sputtering, evaporation, spray deposition, and solution-based sol-gel used to deposit ITO films and reported the role of various deposition parameters on the resultant optical, electrical, and structural parameters [8–10]. However, the demonstrated maximum possible optical transparency around 90% with better electrical conductivity of ITO becomes the landmark limiting value that directly

reduces the solar cell output to $\sim 10\%$ upon the application to solar cell fabrication. In general, in addition to the materials

properties, surface structuring is well in practice to enhance the optical absorbance by reducing the surface impacts on the optical losses of the solar cell [11, 12].

Therefore, in an alternative approach to improve the optical transmittance of ITO films, we tried to establish the surface texturing inherently during the deposition of polycrystalline films

using rf magnetron sputtering. The results are presented here.

[400]

2. Experiment

Indium tin oxide thin films with thickness in the range of 200–300 nm were deposited over glass substrate using the rf power ranging from 25 W to 80 W at an elevated substrate temperature of 250°C in order to maintain the crystallization as well as tin activation [13]. The sputtering was carried out using commercial ITO target composed of 90:10 wt% of In_2O_3 and SnO_2 , respectively. The sputter targeted to substrate distance was retained constant at 5 cm, and the ITO depositions were employed by varying sputtering parameters such as rf power density and the gas composition between Argon and Ar/ O_2 mixture.

Deposited films were characterized using X-ray diffraction analysis, scanning electron microscopy, and UV-visible-NIR spectroscopy. UV-visible spectrophotometer measurements were further used for the analysis of absorption coefficient and optical bandgap properties of the films.

3. Results and Discussion

Structural Characterization. The X-ray diffraction analysis was carried out for the films deposited at different rf powers at a substrate temperature of 250°C. Figure 1 shows the XRD spectra of the films deposited at different rf powers. It is shown that all the films exhibit polycrystalline structure in comparable to the standard JCPDS card number 06-0416 for BCC cubic In_2O_3 (IO) crystal structure except the observed change in the preferred plane of orientation. According to the standard, the basic structure of indium oxide films results in the preferential plane of crystallization along (222) plane, whereas here in Figure 1 it is found that the preferential plane of crystallization is observed as (400) plane. The change in preferential plane is confirmed from the decrease in the $I(222)/I(400)$ value less than 3.33 of the JCPDS report [8]. The value is found to decrease to 0.81 for 30 W and to 0.35 for 80 W, respectively, without major changes in the peak positions. This observation is in analogous to the reported (400) preferential plane of crystallization for the films deposited with poor stoichiometric composition less than 1.5 of In/O ratio. It is reported that the poor oxidation of ITO resulted in poor transparency and better crystallization and conductivity [5].

In order to verify the impact of oxygen stoichiometry on the film crystallization, 3% of oxygen was diluted into the Ar plasma at the deposition power of 30 W. Figure 2, shows the XRD spectra of both films deposited with and without oxygen dilution to the plasma. It can be seen that the oxygen dilution to the Ar plasma resulted in the preferential plane of crystallization comparable to the JCPDS card number mentioned earlier. This oxygen-induced change in the preferential plane of crystallization towards the standard XRD reference states that the ITO crystallization is oxygen stoichiometry-dependent as reported in [8]. The $I(222)/I(400)$ of the film was found to change from 0.81 to 3.94 upon oxygen dilution. This indicates that the oxygen dilution enhances the crystallization of (222) plane stronger than any other plane of crystallization, which may be related to the impact on the type

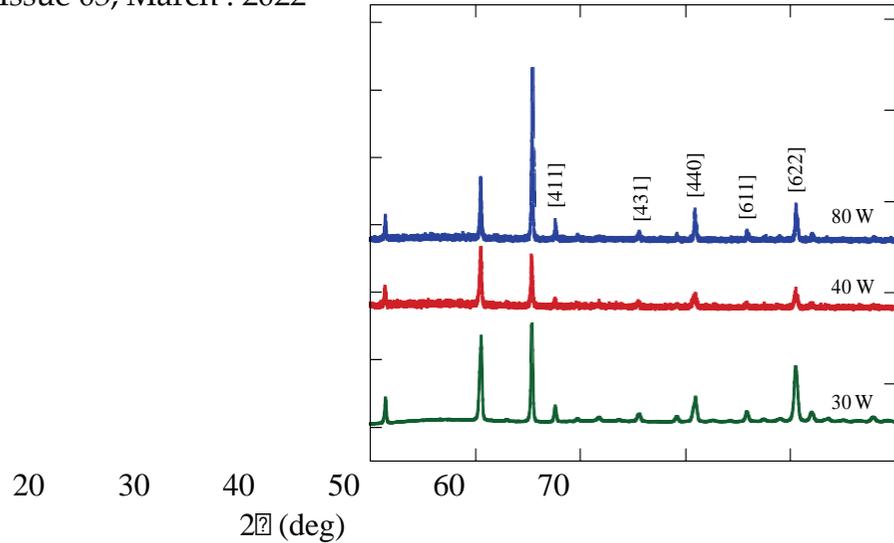


FIGURE 1: X-Ray diffraction pattern of ITO thin film sputtered at different rf powers and substrate temperature of 250°C.

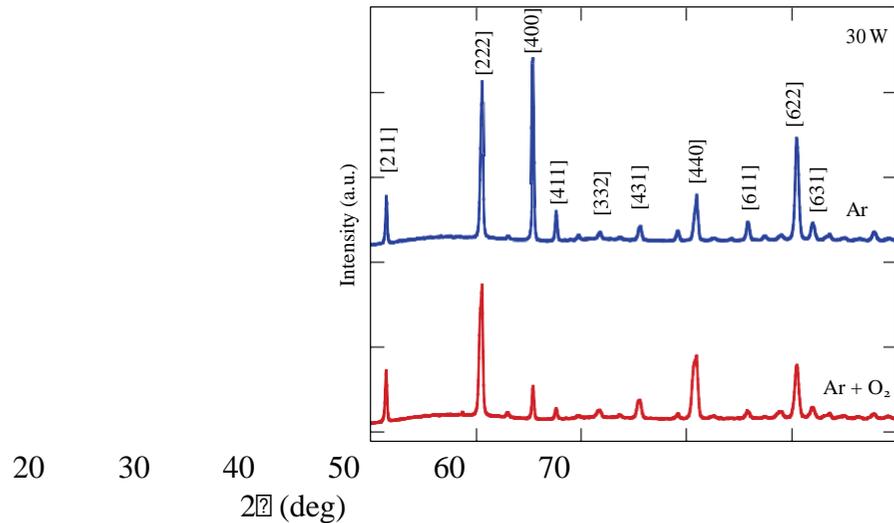


FIGURE 2: X-Ray diffraction pattern of ITO thin film sputtered at 30 W rf power with and without oxygen addition to the plasma.

of oxygen vacancy present in the crystallites, which revoke the habitual crystallization of ITO as reported in [5].

Crystalline sizes of the films were calculated using Scherrer's formula along the preferential plane of crystallizations [14]. The values are found to increase from 39 nm to 52 nm along with the increase in rf power from 30 W to 80 W. This increase in crystalline size along the (400) preferential plane of crystallization clearly indicates the occurrence of columnar growth along this direction of (100). On the other hand, the oxygen dilution and its correlated change in preferential plane of crystallization from (400) to (222) were found to decrease the crystalline size from 29 nm to 21 nm along the (222) direction. Hence, it is identified that the increase in rf power leads to the crystallization of ITO with (400) crystallization, and, on the other hand, the oxygen

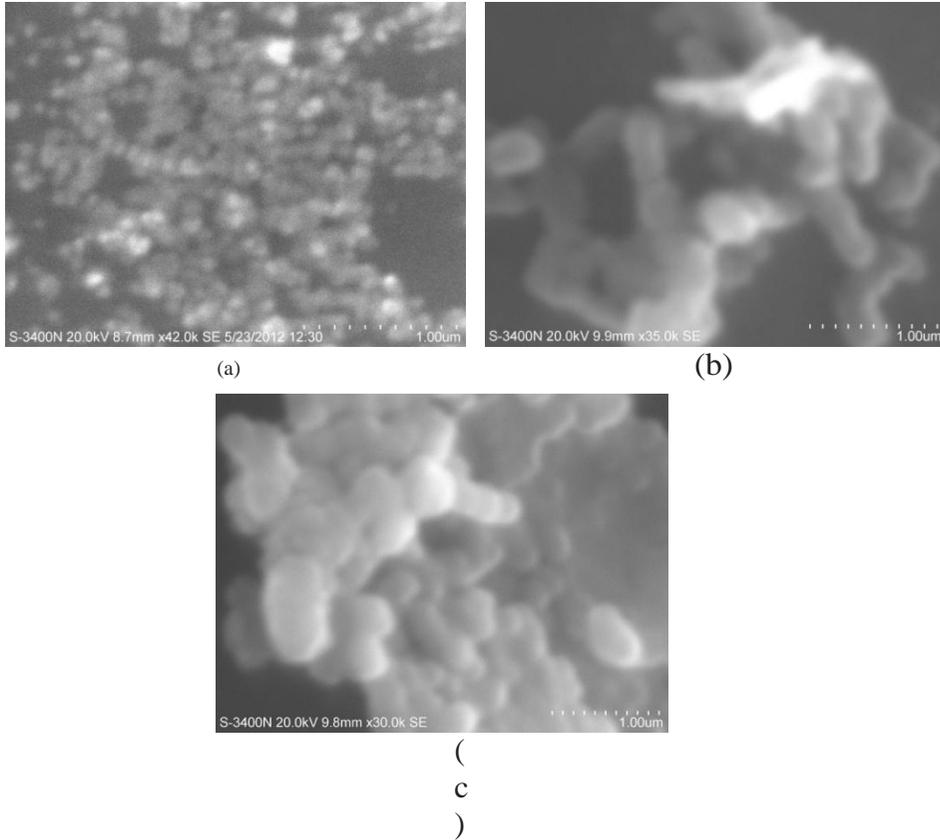


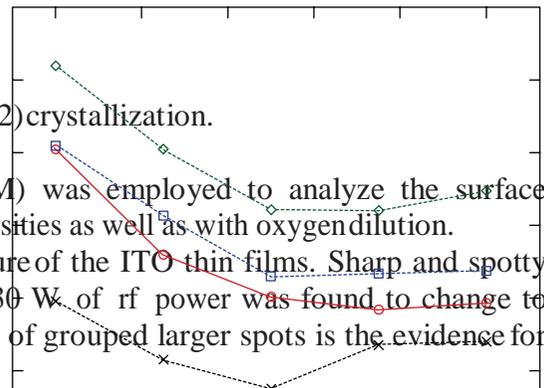
Figure 3: (a) SEM image of the ITO film sputtered at rf power of 30 W at $T = 250^\circ\text{C}$. (b) SEM image of the ITO film sputtered at rf power of 80 W and T of 250°C . (c) SEM image of the film deposited using 30 W and oxygen dilution.

dilution leads to the crystallization of ITO films with (222) crystallization.

SEM Analysis. Scanning electron microscope (SEM) was employed to analyze the surface structure of the films deposited at different rf power densities as well as with oxygen dilution.

Figures 3(a) and 3(b) show the surface microstructure of the ITO thin films. Sharp and spotty surface structure observed for the film deposited using 30 W of rf power was found to change to the grouped few larger spots. This observed less number of grouped larger spots is the evidence for the presence of high value of RMS roughness.

The SEM image of the oxygen diluted film with (222) orientation is shown in Figure 3(c). The figure shows that the films are having granular type of morphology. Hence, it is observed that the change in the preferential plane of



2.8

2.6

2.4

Refractive index



2.2
2
1.8
.6

400 480 560 640 720 800
Wavelength (nm)

crystallization from (400) plane to (222) plane was also found to change the surface microstructure of the rf magnetron sputtered ITO films.

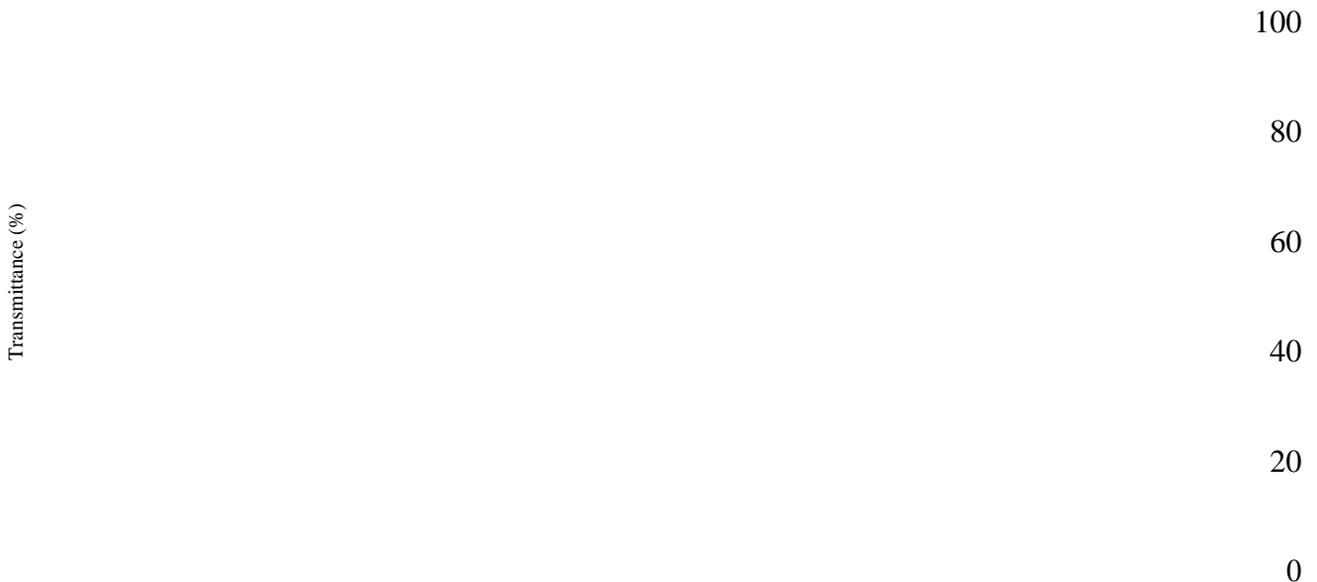
Optical Characterizations. The optical properties such as the transmittance and absorbance spectra were measured using UV-visible spectrophotometer between 300 and 900 nm. It is observed that the increase in rf power from 30 W to 80 W was found to decrease the overall optical transmittance from 77.5% to 60%. The refractive index of the film was calculated using the relation given by [15], and Figure 4



FIGURE 4: Plot wavelength-dependent refractive index of the films deposited at different rf powers.

shows the plot of wavelength-dependent refractive index of the films deposited using different rf powers.

The lowest rf power of 30 W is found to result in the highest transmittance as well as lowest refractive index.



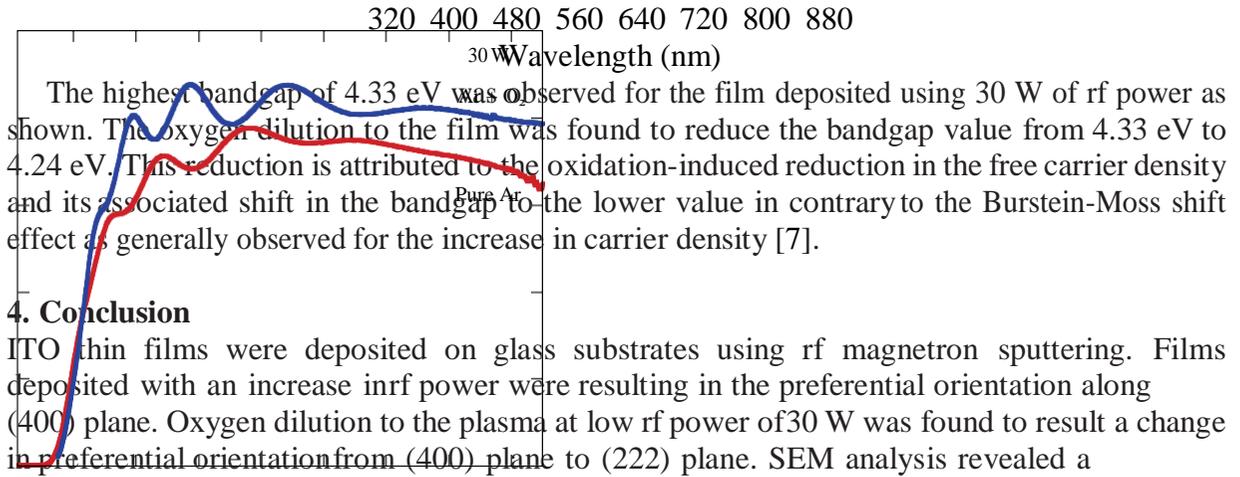


FIGURE 5: Optical transmittance spectra of ITO thin film sputtered using 30 W of rf power and oxygen dilution at a \square of 250°C.

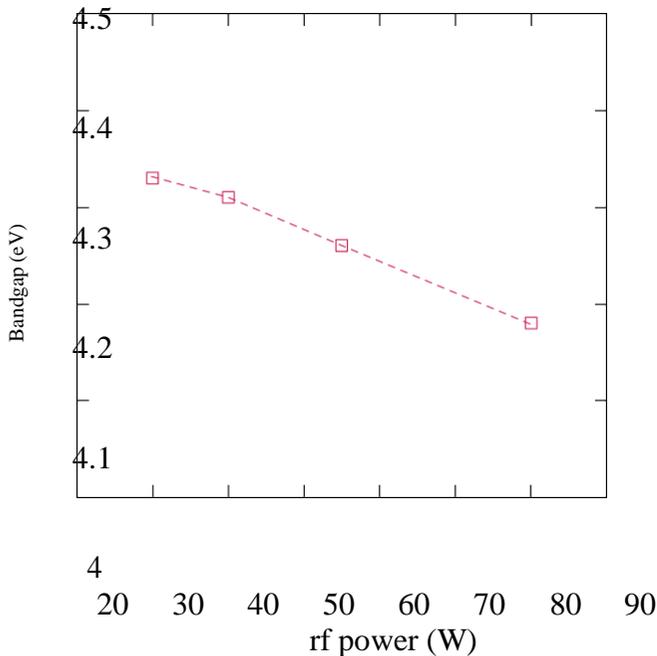


FIGURE 6: Plot of the bandgap values with respect to the rf power used for the deposition.

Figure 5 shows the comparative plot of the transmittance spectra of the films deposited without and with oxygen dilution at the rf power of 30 W. The film deposited with pure Ar plasma is found to result

in the maximum transmittance of 77.5%. Whereas the film deposited with the same power and oxygen dilution was found to result in the transmittance of 87.58%. Hence, it is clear that the oxygen dilution enhances the transmittance value in addition to the change in the preferential plane of crystallization.

Further, absorption coefficient values are calculated using the relation given by Meng and Santos [16]. Calculated values were further used to plot and analyze the band structure properties of the films. Figure 6 shows the plot of the calculated bandgap values with respect to their rf powers. spotty surface morphology for the (400) films and a granular morphology the (222) films. The highest transmittance was found for the oxygen diluted (222) film. Highest optical bandgap of 4.33 eV was observed for the pure Ar deposited film and 4.24 eV for the oxygen diluted film. Therefore, the deposition parameters of rf power and the oxygen dilution are able to result in the change in the film properties as well as surface microtexture.

Acknowledgments

The authors are thankful to the Centre for Nanoscience and Technology and Central Instrumentation Facility (CIF) of Pondicherry University for their support of measurements.

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