TRANSACTIONS ON ELECTRICAL AND ELECTRONIC MATERIALS
Vol. 17, No. 6, pp. 351-354, December 25, 2016

Regular Paper

pISSN: 1229-7607 eISSN: 2092-7592
DOI: http://dx.doi.org/10.4313/TEEM.2016.17.6.351
OAK Central: http://central.oak.go.kr

Study on the Structural and Mechanical Characteristics of ITO Films Deposited by Pulsed DC Magnetron Sputtering

Junyoung Kang, Anh Huy Tuan Le, Hyeongsik Park, Yongjun Kim, and Junsin Yi†
College of Information and Communication Engineering, Sungkyunkwan University, Suwon 16419, Korea

Sunbo Kim
Department of Energy Science, Sungkyunkwan University, Suwon 16419, Korea

Received April 26, 2016; Revised June 16, 2016; Accepted June 20, 2016

The mechanical properties of ITO films such as adhesion and internal stress are very important for the commercial application of solar cell devices. We report high quality pulsed DC magnetron sputtered ITO films deposited on silicon and glass substrates with low resistivity and high transmittance for various working pressures ranging from 0.96 to 3.0 mTorr. ITO films showed the lowest resistivity of 2.68×10^{-4} Ω·cm, high hall mobility of 46.89 cm^2/V.s, and high transmittance (>85%) for the ITO films deposited at a low working pressure of 0.99 mTorr. The ITO films deposited at a low working (0.96 mTorr) pressure had both amorphous and polycrystalline structures and were found to have compressive stress while the ITO films deposited at higher temperature than 0.99 mTorr was mixture of amorphous and polycrystalline and was found to have tensile stress.

Keywords: ITO (indium tin oxide), Working pressure, Internal stress, Pulsed DC magnetron sputtering, SHJ solar cell

1. INTRODUCTION

ITO (indium tin oxide) films are widely used as front TCO (transparent conductive oxide) in LCDs (liquid-crystal displays), OLEDs (organic light emitting devices), and solar cells due to their low resistivity, high transmittance in the visible wavelength region, and wide optical band gap [1-4]. The wide application of ITO films have resulted in an extensive study, both on their preparation and characterization. SHJ (silicon heterojunction) solar cells are being intensively investigated due to their stability, low cost, and low temperature deposition process of amorphous silicon (a-Si) coupled with the high efficiency of crystalline silicon (c-Si). The electrical and mechanical properties of ITO films need to be improved for future highly efficient and high-quality large area SHJ solar cell applications. The mechanical properties of IT films such as internal stress and adhesion are closely associated with the microstructure and deposition process conditions [5,6].

Jung et al. reported the mechanical and structural properties of high temperature a-ITO:Sm films deposited on a polyimide substrate by DC magnetron sputtering [6]. Chuang et al. reported the effect of the working pressure and sputtering power on the properties of ITO flms prepared by long-throw rf magnetron sputtering [7]. The mechanism of stress by sputter deposited metal films was reported by Thornton et al. It was proposed that the atomic peening model of energetic working gas atoms reflected at the cathode for the production of compressive stress at low working gas pressures [12]. Even though several reports have been presented on the mechanical characteristics of ITO films, the lattice strain and binding energy of highly conductive ITO films for SHJ solar cells have rarely been reported.

In this paper, we report the influence of a range of working pressures (0.96 to 3.00 mTorr) on the mechanical properties of highly conductive and transparent pulsed DC magnetron sputtered ITO films. The electrical, optical, and structural properties of ITO films are discussed as a function of working pressure. The

---

† Author to whom all correspondence should be addressed:
E-mail: yi@skku.ac.kr

lattice strain, lattice distance, and binding energy of ITO films are explained for various working pressures.

2. EXPERIMENTAL DETAILS

ITO films were deposited on Corning Eagle XG glass by using a pulsed DC magnetron sputtering system, with an induced power density of 1.23 W/cm^2 and duty cycle of 30%. ITO (100 nm) films were deposited from a sintered ceramic ITO target (8 inch) that was composed of 90 wt% and 10 wt% SnO_2 with 99.999% purity. The substrate temperature, Ar flow rate, and target-substrate (T-S) distance were kept constant at 200℃, 30 sccm and 14.5 cm, respectively, by using a turbo molecule pump. The chamber of the magnetron sputtering unit was evacuated at the pressure of 0.98 mTorr prior to deposition. The working pressure (0.96 to 3.0 mTorr) was controlled by a throttle valve. Spectroscopic ellipsometry (SE VASE®, J. A. Woollam, 240 nm<λ<1,700 nm) was used to measure the thickness of the ITO films. The electrical properties such as the resistivity, Hall mobility, and carrier concentration of ITO films were measured by using the Hall Effect (Ecopia HMS-3000) system at room temperature under a magnetic field of 0.51 T. The optical properties were measured using an UV (ultraviolet)-VIS (visible) spectrophotometer (Scinco S-3100). The crystal structures of the ITO films were characterized from an X-ray diffraction (XRD, D8 Advance, Bruker Inc.) system using CuKα radiation (λ = 0.1541 nm). The XPS (X-ray photoelectron spectroscopy) measurements were carried out using a Thermo VG Scientific Escalab 250 system with a monochromatized Al*K*α (1486.6 eV) X-ray anode at 200 W and 15 kV.

3. RESULTS AND DISCUSSION

Figure 1 shows the XRD patterns of pulsed DC magnetron sputtered ITO (~100 nm) films for various working pressures. The ITO films deposited at low working pressures of 0.96 and 0.99 mTorr were found to have a mixture of In_2O_3 polycrystalline structure, while the films deposited at higher working pressures were found to have an amorphous structure. For higher working pressures of 2.50 and 3.0 mTorr, the kinetic energy of the sputtered particle reaching the surface of the substrate decreased due to high collision frequency between the sputtered particles and gas molecules. The amorphous nature of the ITO films at high working pressures showed an agreement with the already reported work by Sasabayashi and Song et al. [10,13].

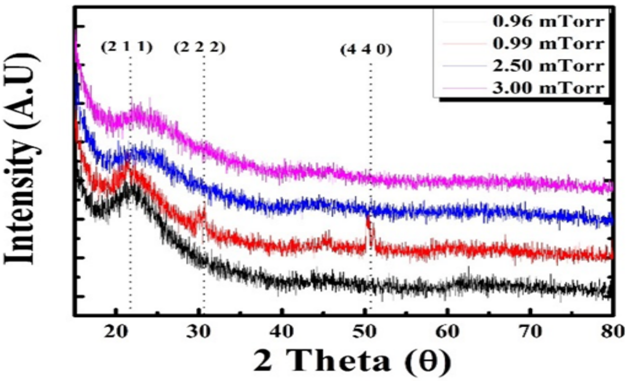

Fig. 1. XRD (X-ray diffraction) patterns of ITO films deposited at various working pressures.

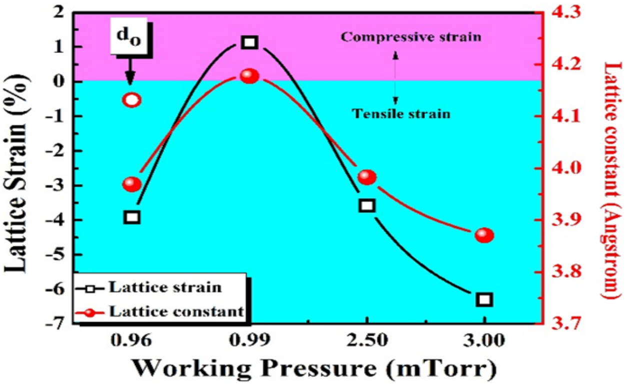

Fig. 2. Lattice strain and lattice constant of ITO films deposited at various working pressures.

From the XRD patterns, we calculated the lattice strain that was strongly related to the off-stoichiometry, point defects, stacking faults, dislocations, etc. Figure 2 shows the lattice strain and lattice constant of the ITO films as a function of working pressure. The ITO films deposited at 0.96 mTorr showed compressive stress, whereas the ITO films deposited from 0.99 to 3.0 mTorr were found to have tensile stress. A positive value of the local strain indicates compressive strain occurring inside the sputtered ITO films. On the other hand, a negative value of the lattice strain showed the tensile strain of ITO films. For high quality ITO films, the compressive or tensile strain should be minimized. Micro cracks at the surface of films can develop with a tensile strain of high value. Lattice strain with a lower absolute value implies that a better collection of charge carrier was obtained. Such large compressive strain in the films deposited at a low working pressure is believed to be caused by the atomic peening effect [11] of high energy neutrals (Ar^0) recoiled from the target or high energy negative ions (O^-) accelerated in the cathode sheath toward the surface of the growing films. The kinetic energies of Ar^0 or O^- decreased due to the increase in collision caused by the increasing working pressure and collision process between the target and the substrate before reaching the substrate surface. Due to high working pressure, all of the Ar^0 reflected at the cathode should be reduced to thermal energy before reaching the substrate [11-13].

The sheet resistance and electrical properties of ITO films (i.e. resistivity (ρ), Hall mobility (μ), and carrier concentration (n)) are shown in Fig. 3 as a function of working pressure. The sheet resistance of ITO films gradually decreased from 54 to 26.82 Ω/sq as the working pressure was increased from 0.96 to 0.99 mTorr.

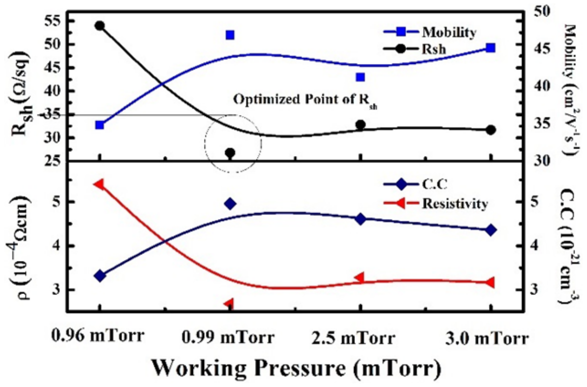

Fig. 3. Sheet resistance and electrical (ρ, n, μ) characteristics of ITO films as a function of working pressure.

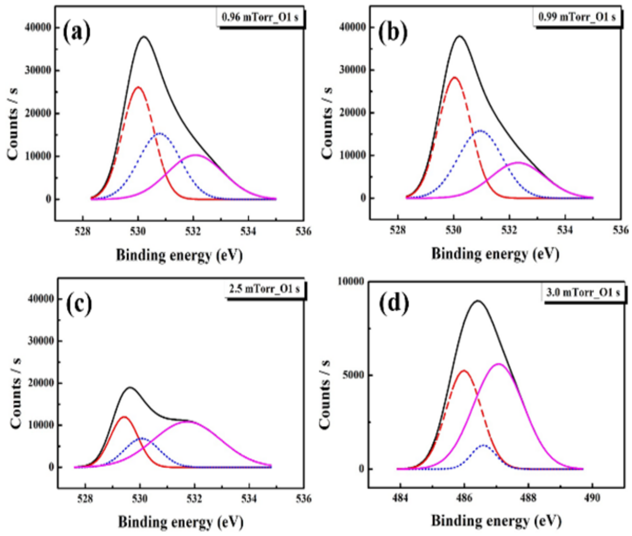

Fig. 4. Binding energy of ITO films deposited with various working pressures i.e. (a) 0.96 mTorr, (b) 0.99 mTorr, (c) 1.5 mTorr, and (d) 3.0 mTorr.

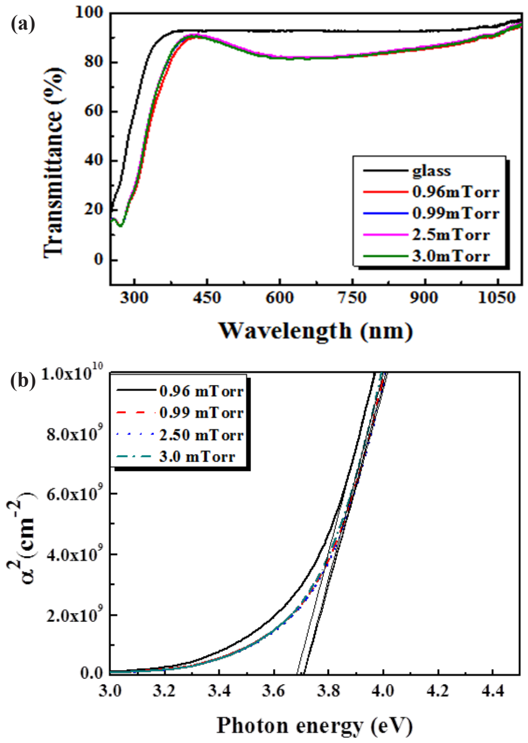

Fig. 5. (a) Optical transmittance and (b) bandgap of ITO films deposited at various working pressures.

With the further increase of working pressure from 0.99 to 3.0 mTorr, the sheet resistance of ITO films increased to 31.69 Ω/sq. The resistivity of ITO films decreased from 5.4×10^{-4} to 2.682×10^{-4} Ω·cm with the increase of working pressure from 0.96 to 0.99 mTorr. With the further increase of working pressure up to 3.0 mTorr, the resistivity of ITO films increased up to 3.169×10^{-4} Ω·cm. The decrease in resistivity of the ITO films is related to the combined influence of the Hall mobility and carrier concentration [14-16]. The maximum value of the Hall mobility of ITO films was recorded as 46.89 (cm^2/V.s) for the working pressure of 0.99 mTorr. The working pressure showed a minor influence on the carrier concentration of ITO films.

To understand the oxygen states and their distribution in the films, we characterized the XPS (X-ray photoelectron spectroscopy) spectra of the ITO films for various working pressures as shown in Fig. 4. The O1s spectra were fitted with the energy components at ~529.6 eV, ~531.2 eV, and 532.0 eV by using three Gaussian functions of variable positions, widths, and intensities. The results agreed well with those reported in the literature [17,18]. The $O_{529.6}$ and $O_{531.2}$ peaks originate from the In_2O_3 regions and the oxygen deficient regions respectively, while the peak of $O_{532.0}$ mainly originates from oxygen contamination [17,18].

The optical transmittance of the ITO films as a function of working pressure is shown in Fig. 5(a). Bare Eagle glass showed 92% transmittance in the visible wavelength (400~800) nm region. All the ITO films showed more than 85% transmittance in the vis-NIR (400~1,100) nm wavelength region [1-5,20]. The optical bandgap of ITO films was obtained by extrapolating the linear part on the hν axis of the plot $(\alpha h\nu)^2$ vs. hν as shown in Fig. 5(b) for various working pressures. The ITO films deposited at the working pressure of 0.96 mTorr showed the optical bandgap of 3.69 eV while all the other ITO films showed an optical bandgap of above 3.705 eV.

4. CONCLUSIONS

Highly transparent and conductive pulsed DC magnetron sputtered ITO films deposited on silicon and glass substrates are reported for various working pressures ranging from 0.96 to 3.0 mTorr. Among the ITO films, those deposited at the working pressure of 0.99 mTorr showed the lowest sheet resistance of 26.82 Ω/sq, resistivity of 2.68×10^{-4} Ω·cm, Hall mobility of 46.89 cm^2/V.s, and a total transmittance of 85.94%.We achieved a lower absolute value of lattice strain for ITO films, indicating that a better collection of charge carrier was obtained. The XRD analysis showed the polycrystalline nature of ITO film deposited at a low working pressure. Due to their excellent electrical and mechanical properties, pulsed DC magnetron sputtered ITO films are proposed for future high efficiency SHJ solar cell.

REFERENCES

[1] V. A. Dao, H. Choi, J. Heo, H. Park, K. Yoon, Y. Lee, Y. Kim, N. Lakshminarayan, and J. Yi, *Curr. Appl. Phys.*, **10**, S506 (2010). [DOI: http://dx.doi.org/10.1016/j.cap.2010.02.019]

[2] W. K. Oh, S. Q. Hussain, Y. J. Lee, Y. Lee, S. Ahn, and J. Yi, *Mater. Res. Bull.*, **47**, 3032 (2012). [DOI: http://dx.doi.org/10.1016/j.materresbull.2012.04.106]

[3] S. Kim, J. Jung, Y. J. Lee, S. Ahn, S. Q. Hussain, J. Park, B. S. Song, S. Han, V. A. Dao, J. Lee, and J. Yi, *Mater. Res. Bull.*, **58**, 83 (2014). [DOI: http://dx.doi.org/10.1016/j.materresbull.2014.05.003]

[4] S. Q. Hussain, J. H. Lee, S. Kim, S. Ahn, N. Balaji, Y. Lee, and J. Yi, *Sol. Energy Mater. Sol. Cells*, **122**, 130 (2014). [DOI: http://dx.doi.org/10.1016/j.solmat.2013.11.031]

[5] S. Q. Hussain, W. K. Oh, S. Ahn, A.H.T. Le, S. Kim, Y. Lee, and J. Yi, *Vacuum*, **101**, 18 (2014). [DOI: http://dx.doi.org/10.1016/j.vacuum.2013.07.004]

[6] T. D. Jung and P. K. Song, *Curr. Appl. Phys.*, **11**, S314 (2011). [DOI: http://dx.doi.org/10.1016/j.cap.2011.03.042]

[7] M. Chuang, *J. Mater. Sci. Technol.*, **26**, 577 (2010). [DOI: http://dx.doi.org/10.1016/S1005-0302(10)60088-6]

[8] S. H. Chang, H. M, Cheng, C. L. Tien, S. C. Lin, and K. P. Chuang, *Optic. Mater.*, **38**, 87 (2014). [DOI: http://dx.doi.org/10.1016/

j.optmat.2014.10.004]

[9] R. X. Wang, C. D. Beling, A. B. Djuriši, S. Li, and S. Fung, *Semicond. Sci. Technol.*, **19**, 695 (2004). [DOI: http://dx.doi.org/10.1088/0268-1242/19/6/006]

[10] P. K. Song, Y. Shigesato, I. Yasui, C. W. O. Yang, and D. C. Paine, *Jpn. J. Appl. Phys.*, **37**, 1870 (1998). [DOI: http://dx.doi.org/10.1143/JJAP.37.1870]

[11] S. Takayama, T. Sugawara, A. Tanaka, and T. Himuro, *J. Vac. Sci. Technol.*, **A21**, 1351 (2003). [DOI: http://dx.doi.org/10.1116/1.1563623]

[12] J. A. Thronton and D. W. Hoffman, *Thin Solid Films*, **171**, 5 (1989). [DOI: http://dx.doi.org/10.1016/0040-6090(89)90030-8]

[13] T. Sasabayashi, N. Ito, E. Nishimura, M. Kon, P. K. Song, K. Utsumi, A. Kaijo, and Y. Shigesato, *Thin Solid Films*, **445**, 219 (2003). [DOI: http://dx.doi.org/10.1016/j.tsf.2003.08.047]

[14] K. Utsumi, H. Iigusa, R. Tokumaru, P. K. Song, and Y. Shigesato, *Thin Solid Films*, **445**, 229 (2003). [DOI: http://dx.doi.org/10.1016/S0040-6090(03)01167-2]

[15] A. Klein, C. Korber, A. Wachau, F. Sauberlich, Y. Gassenbauer, S. P. Harvey, D. E. Proffit, and T. O. Mason, *Materials*, **3**, 4892 (2010). [DOI: http://dx.doi.org/10.3390/ma3114892]

[16] S. Q. Hussain, W. K. Oh, S. Kim, S. Ahn, A.H.T. Le, H. Park, Y. Lee, V. A. Dao, S. Velumani, and J. Yi, *J. Nanosci. Nanotechnol.*, **14**, 9237 (2014). [DOI: http://dx.doi.org/10.1166/jnn.2014.10142]

[17] S. Ahn, S. Kim, V. A. Dao, S. Lee, S. M. Iftiquar, D. Kim, S. Q. Hussain, H. Park, J. Lee, Y. Lee, J. Cho, S. Kim, and J. Yi, *Thin Solid Films*, **546**, 342 (2013). [DOI: http://dx.doi.org/10.1016/j.tsf.2013.05.132]

[18] S. Kim, J. Lee, V. A. Dao, S. Lee, N. Balaji, S. Ahn, S. Q. Hussain, S. Han, J. Jung, J. Jang, Y. Lee, and J. Yi, *Mater. Sci. Eng. B*, **178**, 660 (2013). [DOI: http://dx.doi.org/10.1016/j.mseb.2012.10.029]

[19] E. H. Kim, C. W. Yang, and J. W. Park, *J. Appl. Phys.*, **109**, 043511 (2011). [DOI: http://dx.doi.org/10.1063/1.3556452]

[20] W. F. Wu and B. S. Chiou, *Appl. Surf. Sci.*, **115**, 96 (1997). [DOI: http://dx.doi.org/10.1016/S0169-4332(96)00852-5]