

Effects of Laser Radiation on the Optical and Electrical Properties of ITO Thin Films Deposited by RF Sputtering

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Abstract Annealing treatment of transparent conducting oxide (TCO) thin films plays a great role in enhancing the optoelectronic properties of the material. Changes in morphological, optical and electrical properties of indium tin oxide (ITO) thin films deposited by RF sputtering were investigated after exposing the films to Nd:YAG laser radiation. ITO thin films of 158 nm thickness were irradiated with different laser energy; 25 mJ, 75 mJ, 120 mJ and 165 mJ respectively. Atomic force microscopy (AFM) results reveal a smooth surface morphology and enhance grain size as the laser energy increases. Highest optical transmittance value of 96.5 % at 620 nm wavelength was obtained by film treated with 165 mJ laser energy as determined by UV-Vis spectrophotometer. Electrical resistivity measurements as determined by four-point probe show a significant decrease in resistivity and sheet resistance with respect to increasing laser energies. The ITO films optoelectronics properties were enhanced with the film annealed at 165 mJ exhibiting the highest calculated figure of merit. This laser treatment method has effectively fine turned the ITO films properties toward TCO functional properties required for solar cell application.

Keywords ITO, Laser-annealing, Nd:YAG Laser, RF Sputtering, Solar Cell, TCO

1. Introduction

Incorporation of transparent conducting oxides (TCO) thin layers in silicon (Si) solar cells is one of the progresses so far achieved in the development of photovoltaic devices. TCO layers serve as an anti-reflection coating (ARC),

junction rectifiers and transparent electrode for the extraction of separated charge carriers from the absorbing region of semiconductor material[1-3]. Other applications like organic light-emitting device (LED)[4] and flat panel display (FPD) [5] also employed TCO films as transparent conductive contact. Different TCO thin films materials have been used in this regard, such as zinc oxide (ZnO), gallium-doped indium tin oxide (GITO) and indium tin oxide (ITO) [6-8]. ITO thin film is the most widely applied TCO due to its excellent electrical conductivity ($10^4 \Omega^{-1} \text{cm}^{-1}$) and high optical transmittance ($> 80 \%$) in the visible region of light spectrum[9, 10]. It's a degenerate n-type semiconductor with an increasingly wide bandgap (3.5-5.1 eV) [11, 12]. ITO high transparency is as a result of its wide bandgap, whereas the presence of oxygen vacancies and extrinsic defect of tin dopant contributed to its high electrical conductivity [3, 13, 14].

Deposition technique plays a key role in obtaining an improved optical and electrical properties of ITO thin films. Various methods of thin films deposition have been used: chemical vapour deposition method [12]; spray pyrolysis[13]; electron beam evaporation [14]; sol-gel method[15]; and direct current (DC) or RF magnetron sputtering technique [19,20]. The production of uniform and excellent quality thin films with good adherence to a different type of substrates by RF sputtering has often encouraged the use of this method by the researchers [14, 21, 22]. Apart from the deposition technique, temperature annealing, films thickness and operating parameters also contributed to the changes in the optoelectronic properties of this material [23–26].

RF Sputtered ITO films at room temperature resulted in amorphous nature. Annealing is required to improve the structural crystallization and electrical conductivity in order to get the needed functionality of the thin films.

Alternatively, laser annealing can be used to enhance thin films structural crystallization due to the effect of laser radiation on the material structural and morphological properties[24]. The optoelectronic properties of the ITO films can be enhanced by the application of high energy pulsed laser radiation that impacted positively on the localised surface region of the thin films during recrystallization process [27-29].

In this work, we present the effect of pulsed laser annealing on ITO thin films sputtered deposited on n-type silicon. Solid-state Nd:YAG pulsed laser with 1064 nm wavelength was used as a radiation source. The samples were characterised based on surface morphological, optical and electrical properties.

2. Materials and Methods

ITO ceramic target (90 % In_2O_3 and 10 % SnO_2) was used to deposit ITO thin films on Si and glass substrates using radio frequency (RF) magnetron sputtering system. Before the deposition process, Si substrates were cleaned in acetone and isopropyl alcohol (IPA) to remove surface impurities, and finally rinsed in deionised (DI) water and blown dry using N_2 gas. Decon-90 glass cleaner and acetone were used to clean the corresponding glass substrates and then rinsed in DI water and subsequently blown dry under N_2 gas. Prior to ITO films deposition, the sputtering chamber was evacuated to 8.9×10^{-6} Torr, and then, the chamber was filled back again with Ar gas at a flow rate of 50 sccm. Target to substrate distance was kept at 130 mm while working pressure and RF power were set at 5.24×10^{-3} mTorr and 120 W respectively. A deposition time of 10 min was set and before that, a pre-sputtering process was conducted for 15 mins to clean the target surface from the contaminant particles. All the process took place at room temperature.

As a source of annealing energy, a Neodymium-doped Yttrium Aluminium Garnet (Nd: YAG) laser Series (NANO L290-20) by Litron with 1064 nm fundamental wavelength and 4 ns pulse duration was applied as a laser

annealing treatment on ITO thin films surfaces. The pulse energy of 25 mJ, 75 mJ, 125mJ, and 165 mJ was used under N_2 gas atmosphere to radiate the ITO samples. Thickness measurement and morphological characteristics were carried out using the surface profiler and atomic force microscopic (AFM) systems. Ultraviolet-visible (UV-Vis) spectrophotometer and four-point probe systems were used to analyse the optical and electrical properties of the ITO thin films.

3. Results and Discussion

Surface morphology, optical and electrical properties of the as-deposited and irradiated ITO thin films samples were characterized accordingly. One of the important and useful techniques used for the analysis of the surface morphology of thin films materials is atomic force microscopy. Fig. 1 shows three dimensional (3D) AFM images of ITO films with and without Nd:YAG laser energy treatment scanned over $5 \mu\text{m} \times 5 \mu\text{m}$ area. The morphological parameters of the treated and untreated ITO films were tabulated in Table 1. A careful study of these images in Fig. 1 and Table 1 indicates the dependance of ITO films surface morphology on the amount of laser energy exposed on their respective surfaces. The grain size, as well as the respective roughness of the treated samples, increased significantly when the applied laser radiation value is increased due to laser heat absorption. This causes the grain to expand and agglomerate resulting in rougher surface. The large grain size decreases the grain boundary scattering which subsequently improves the carrier mobility of the ITO films. Some larger dots with white spots are seen on the surface of the treated ITO films. Generally, the regular grain growth of morphological features of the treated samples with dips, cavities surface pattern aided the surface roughness[24]. As seen in Table 1 smaller root means square R_q and average R_a roughness value for film treated at 25 mJ laser energy was obtained as compared to 165 mJ and similar result was obtained by Butt et al. [24] and Wang et al.[25].

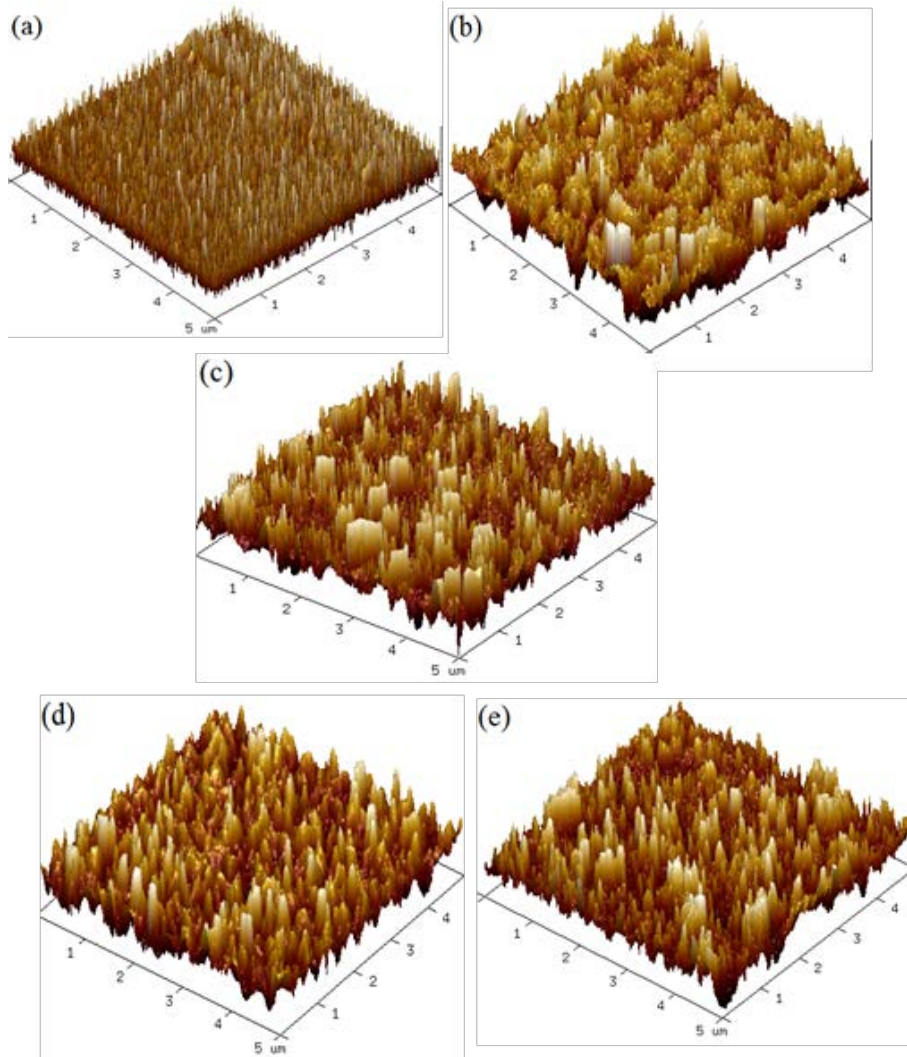


Figure 1. AFM images of ITO films for the (a) as-deposited (b) 25 mJ (c) 75 mJ (d) 120 mJ and (e) 165 mJ

Table 1. ITO root means square (RMS) R_q and average R_a roughness and grain size with respect to increasing laser energy

Factor	0 mJ	25 mJ	75 mJ	120 mJ	165 mJ
RMS R_q (nm)	1.5	5.5	6.2	6.3	10.1
R_a (nm)	0.85	3.65	4.9	5.3	8.5
Grain size (nm)	65.5	76.3	90.3	92.9	97.9

Fig. 2 displays the optical transmittance and absorption spectral features of as-deposited and laser annealed ITO thin films at different energies. From Fig. 2a, the as-deposited film shows the transmittance of 91.8 % at 620 nm. Gradual improvement in transmittance spectra was achieved as the laser energy increases. A maximum transmittance of 96.5 % at 620 nm was obtained when 165 mJ Nd:YAG laser radiation was directed onto the surface of the as-deposited ITO films. The occurrence of high transmittance at this wavelength range is necessary for any electrode to serve as an antireflection coating [1], [26]. The continuous interfering fringes of the laser-treated samples from visible-infrared regions indicate good uniformity in the thin films. A slight decrease in absorption was observed as the value of laser energy increases as shown in Fig. 2b. This shows that the ITO films surface atoms absorption ability gradually reduces when ITO films were irradiated with increasing laser energy. This in return, improved and also allows the smooth passage of radiation intensity through the material.

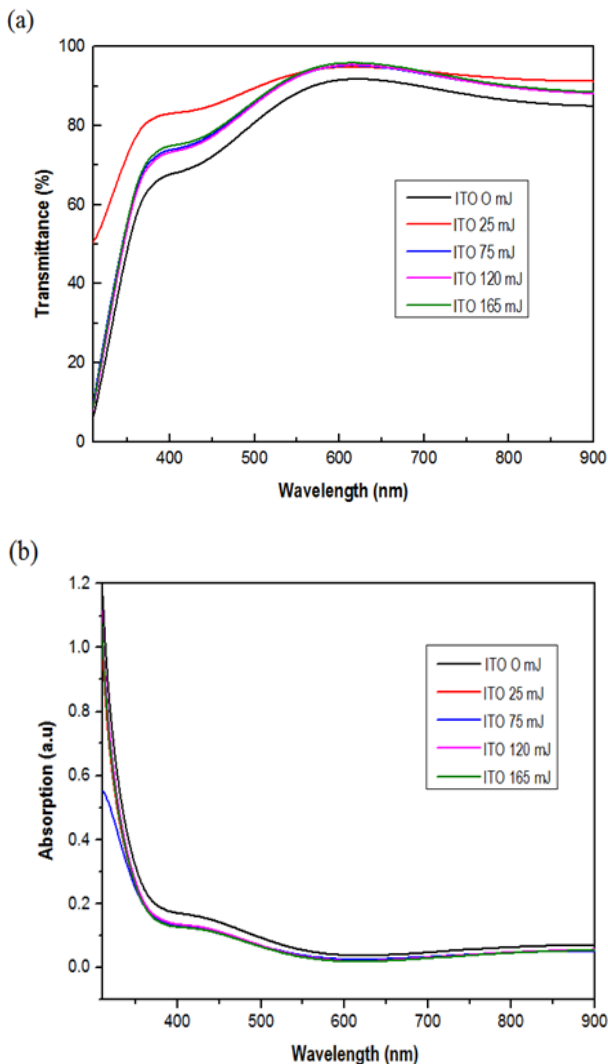


Figure 2. ITO films (a) transmittance spectra and (b) absorption spectra of the as-deposited and laser annealed samples

ITO films electrical properties are shown in Fig.3. As indicated the electrical resistivity decreases gradually as the laser energy increases. An electrical resistivity ρ of $3.69 \times 10^{-4} \Omega\text{-cm}$ with sheet resistance R_{sh} 23.4 Ω/sq was obtained for film annealed at 165 mJ laser energy. The ρ and R_{sh} decrease with respect to increase laser energy radiation. And this is attributed to the existence of oxygen vacancies, dislocation line density increment and laser annealing domination over a defect in the material [27].

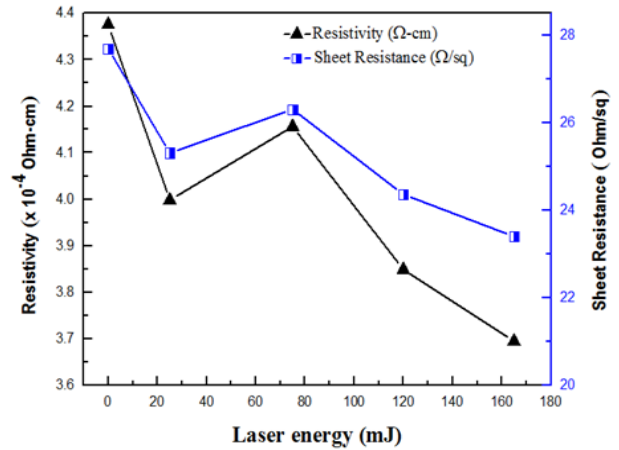


Figure 3. Resistivity and Sheet resistance as a function of laser energy

High optical transmittance and good electrical conductivity are essential for any TCO thin films to perform excellently as a transparent conducting electrode (TCE) in display or photovoltaic devices. The performance of this electrode is evaluated by a figure of merit (FOM). The evaluation of maximum transmittance and FOM value for irradiated ITO films of different laser energy is shown in Fig. 4. Sheet resistance R_{sh} and transmittance T are two parameters that constitute the $FOM = T^{10}/R_{sh}$ value [20]. As seen from Fig.4, the FOM value increased as the laser energy increases from 25 mJ to 165 mJ due to simultaneous enhancement in electrical conductivity and transmittance in the visible range [20].

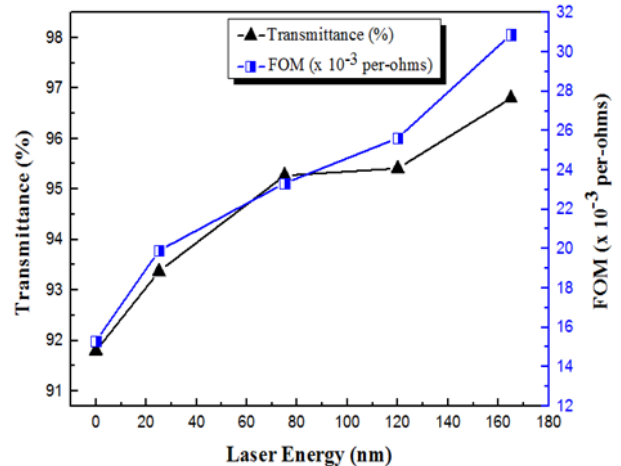


Figure 4. Optical transmittance and FOM as a function of 0 mJ, 25 mJ, 75 mJ, 120 mJ and 165mJ laser energies.

4. Conclusions

Laser annealing treatment of ITO thin films by Nd:YAG laser system has been investigated as an approach to improve the ITO optoelectronics properties. Effects of laser treatment on ITO films based on morphological, optical and electrical properties have been analysed. Grain size and surface roughness increased as the energy of pulsed laser radiation is increased. Maximum transmittance of 96.5 % at antireflection wavelength (620 nm) and resistivity of $3.69 \times 10^{-4} \Omega\text{-cm}$ were obtained by film treated with 165 mJ laser energy. The concurrent improvement in transmittance and conductivity of the treated ITO films as evaluated by FOM shows that Nd:YAG nanosecond pulsed laser annealing can be a promising alternative to temperature annealing. The treated ITO films surface profile, optical transmittance spectra and electrical conductivity properties satisfied some functional properties requires of TCO in solar cells devices.

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