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Tailoring of the Structural and Optoelectronic Properties of Zinc-Tin-Oxide Thin Films via Oxygenation Process for Solar Cell Application

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ABSTRACT In this study, the impact of compositional variation in high resistance transparent (HRT) metal oxide ZTO films of thickness around 100nm has been investigated. The atomic composition in the films has been tailored by the change of RF power and sub-sequent thermal oxygenation in mixed nitrogen and oxygen atmosphere. A phase transition from $ZnSnO_3$ to $ZnSnO_4$ was observed in the X-ray diffraction spectra, indicating the possible oxygen incorporation into the films during the thermal annealing process. Uniform microstructures with compact interconnected grains of around 6-7 nm were found in SEM images while no significant changes been observed upon oxygenation. Besides, the significant alteration of electronic properties was noticed as an effect of compositional variation via oxygenation. All the films showed above 85% of optical transmittance in the visible light spectrum. The optimum optoelectronic properties for RF power has been determined as of 50W (ZnO) and 10W (SnO_2) via thermal oxygenation at 400°C where the ratio O/(Zn+Sn) become around 1.6. The significant effect of oxygenation has been realized via primarily fabricated solar cells where the cell with $ZnSnO_4$HRT shows higher efficiency than the $ZnSnO_3$.

INDEX TERMS ZTO thin film, co-sputtering, phase transformation, HRT film, CdTe solar cell.

I. INTRODUCTION

In recent years, high resistance transparent (HRT) metal oxide compounds have drawn widespread attention for improving the photovoltaic characteristics of thin-film heterostructure solar cells. Particularly, using HRT materials as a buffer layer beneath of the cadmium sulfide (CdS) layers in cadmium telluride (CdTe), copper-indium-gallium-selenide (CIGS), and copper-zinc-tin-sulfide (CZTS) thin-film solar cells can significantly ameliorate the pin-hole problem that is created due to the ultra-thin CdS layer (($\leq$100 nm). In general, the ultra-thin CdS layer is being used in these solar cells for minimizing the considerable absorption by CdS thin film in the blue region [1]–[3]. However, the ultra-thin CdS layer adversely affects the cell efficiency via increasing pin-hole (discontinuity) and locally shorted the absorber material with TCO and front contact, which leads to excessive shunting or unwarranted forward current, and therefore negatively affects the solar cell efficiency [4]. Apart from the reduction of the pin-hole effect, the inclusion of an HRT layer shows improvement on the uniformity and junction quality in a manner parallel to that found for CdTe [5], [6], $CuInSe_2$/CdS, and a-Si thin-film cells [7]. Several materials have already been tested for the HRT layer including the SnO_2 [8], In_2O_3 [9], TiO_2 [10], Ga_2O_3 [11], and Zn_2SnO_4 / $ZnSnO_3$ (ZTO) [12]. Among the aforementioned HRT materials, the ZTO shows

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very promising for solar cell application because of its high carrier mobility (in the range of 5–20 cm^2/Vs) [13], high optical transmittance (above 80% in the visible region), and admirable resistivity (∼10−20 Ω-cm) [14], [15]. Besides, the optoelectronic properties of ZTO films can also be tailored by changing the atomic ratio of oxygen (O) to tin (Sn) by ensuring a proper stoichiometry of the film. Furthermore, the ZTO has high electron affinity (5.3 eV) for which it catches attention as a hole extracting material in polymer and organic solar cells [16]. Likewise, other than the solar cell application, amorphous ZTO thin films have widely been used for flexible field-effect transistors (FETs) [17] and thin-film transistors (TFTs) [18].

There are two types of ZTO: the spinel-type of $Zn_2SnO_4$and the perovskite-type of $ZnSnO_3$. The resistivity of Zn_2SnO_4 is usually higher than that of $ZnSnO_3$ [19]. This is reflected well in literature data of conductivity for ITO ($\sigma = 104\ \Omega^{-1}cm^{-1}$ [20]), for HRT layers as Zn_2SnO_4 (σ = 50-100 $\Omega^{-1}cm^{-1}$ [21, 22]) and for $ZnSnO_3$ ($\sigma = 250\ \Omega^{-1}cm^{-1}$ [17], [23]). It should be mentioned that the properties of ZTO films are critically dependent on the growth conditions. Amended understanding of ZTO film's structural evolution, surface morphology, optical and electrical properties with its growing conditions are indispensably needed not only for the enrichment of all concurrent practical applications but also for reconnoitering the novel possible technology. The primary objective of the present study is to find out the influence of fabrication parameters, especially the oxygenation process on the structural, morphological, and optoelectronic properties of the co-sputtered ZTO films. It should be noted that the sputtering technique could facilitate precise thickness, chosen elemental composition with a controlled amount of impurity in a long-range, thus, it could be the utmost deposition method for ZTO thin film fabrication.

II. METHODOLOGY

A. EXPERIMENTAL DESIGN AND PROCEDURE

ZTO thin films of about 100 nm thickness were deposited by radio frequency (RF) magnetron co-sputtering from ZnO (Plasma Materials 99.999%) and SnO_2 (Plasma Materials 99.999%) targets on commercially available quartz glass substrates. The co-sputtering process is shown schematically in Figure 1(a). The sputtering has been done in Ar ambient with the total pressure to be 8-10mT, with baseline pressure of 10^{-2} mT. The films were deposited at 300°C of substrate temperature using deposition RF power of 50 watts for ZnO, and 05 watts and 10 watts for SnO_2 target. The deposition power in the SnO_2 target was varied to obtain the film with different Zn/Sn ratios. The extra oxygen was induced in the films via thermal annealing in N_2 and O_2 ambient with ratio (80:20) at two high temperatures 400°C and 500°C for 30 minutes. A simple quartz tube has been used for the oxygenation process as shown schematically in Figure 1(b) which has a gas inlet, a vacuum outlet, two electrical coils (one on top and another at the bottom), each of 2 kW.

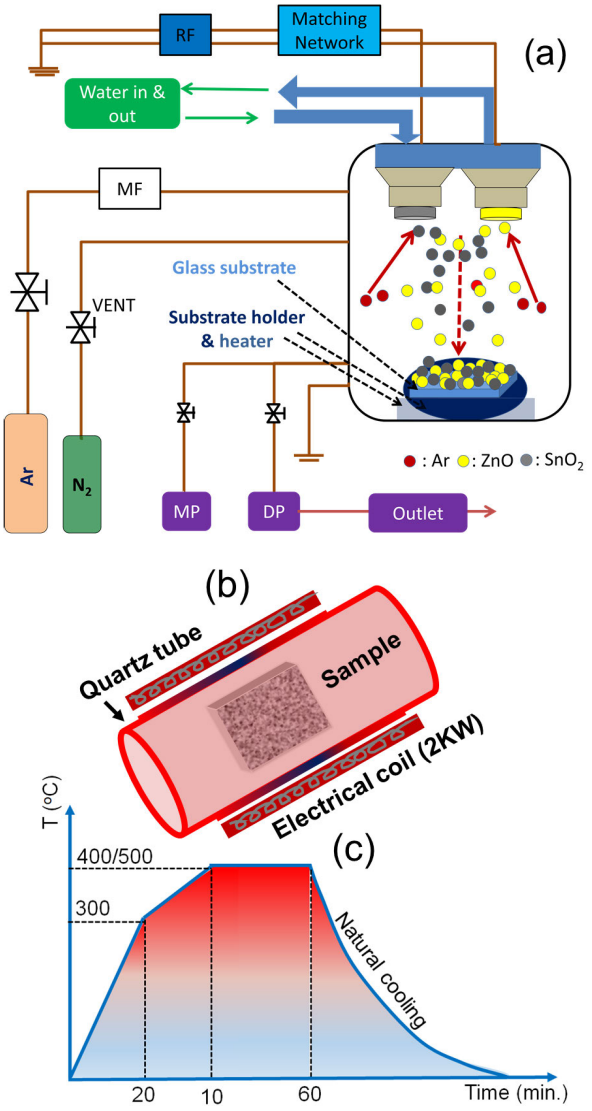

FIGURE 1. (a) Schematic diagram of the co-sputtering process, (b) Schematic diagram of the thermal annealing process, and (c) thermal annealing profil.

B. CHARACTERIZATION

X-ray diffraction (XRD) spectroscopy ('BRUKER aXS-D8 Advance Cu- Kα) has been employed for investigating the structural variation in the films. The surface morphology of the films was observed from the FESEM images carried out by 'LEO 1450 Vp'. Electronic properties including carrier mobility, concentration, and resistivity of the films were measured using the Hall-Effect measurement system 'ECOPIA 3000'. The film transmittance, absorbance, and optical bandgap were measured by using Perkin Elmer Instruments Lambda35 UV-vis spectrometry.

The complete solar cells have been primarily fabricated using ZTO thin films as HRT material that deposited on top of the commercial fluorine-doped tin oxide (FTO) coated glass substrates. The subsequence CdS and CdTe thin films have also been fabricated on top of the FTO/ZTO stacks. The cell fabrication was completed using

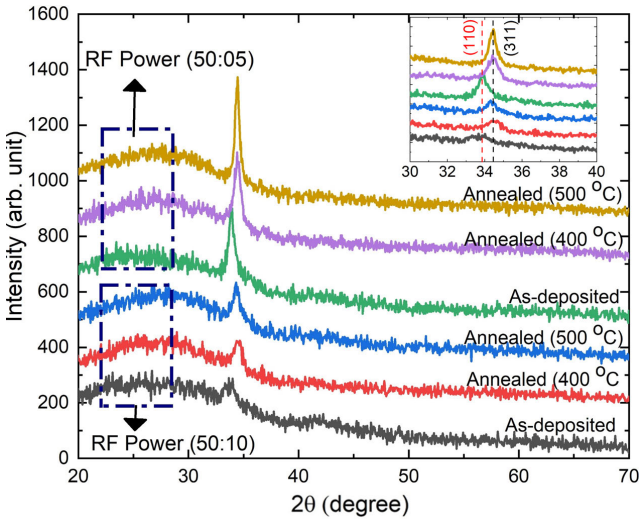

FIGURE 2. XRD diffraction spectra of co-sputtered ZTO thin films prepared using different RF power and subsequent oxygenation in N_2/O_2 ambient (peaks are assigned by JCPDS No. 010890095 for $ZnSnO_3$ and 010731725 for Zn_2 SnO_4).

C: Cu/Ag back contact. Before fabricating the back-contact, the Glass/FTO/ZTO/CdS/CdTe stacks were $CdCl_2$ treated for achieving Te rich surface. The printed paste was prepared by 3 gm of Cu powder adding to the 10 gm of commercial Carbon paste and mixed them for 1 h. using a rotating magnet. The C: Cu printed layers were dried for 30 min at a temperature of 120 °C in an oven. After that, the completed cells were annealed for 15 min in a vacuum furnace at 500 mT of N_2 pressure and temperature of 260 °C. During the vacuum annealing, Cu supposed to be diffused and expecting to be formed an ultra-thin Cu_xTe layer on the CdTe surface. Finally, device fabrication was completed by printing Ag as a second electrode. The performance of the cell with an area of 25 mm^2 was evaluated under standard illumination (1.5 AM) employing 'Gratings Inc. Solar Cell Tester: VI & power management system'.

III. RESULTS AND DISCUSSION

A. STRUCTURAL ANALYSIS

The XRD analysis was performed to investigate the crystallographic growths including crystallinity and crystal phases of the prepared ZTO films as shown in Figure 2. The broad peaks with very low intensity indicating poor crystallinity. On the other hand, there is a peak shift from (110) of $ZnSnO_3$ (JCPDF, card no. 24-1470) to (311) of Zn_2SnO_4 (JCPDS file No: 01-074-2184) has been observed due to the annealing in an oxygen ambient. Peak shifting confirms that additional oxygen atoms have been incorporated into the ZTO lattice during the annealing and shifting occurred to the higher angle because the oxygen ions (O^{2-}) have smaller radii than Zn^{2+} ions and Sn^{4+} ions. Also, due to the smaller ionic radii, the oxygen ions easily substitute the Zn and/or Sn ions. Similar phenomena could also occur if Zn ions are replaced by Sn ions [24]. It has been identified that the amorphous structure of ZTO is dominant instead of a crystalline

TABLE 1. Estimated structural properties of the co-sputtered ZTO thin films and subsequent oxygenation via thermal annealing in N_2: O_2 ambient.

RF Power (W)	Sample ID	2θ (°)	Plane (hkl)	I (a.u.)	D (nm)	ε ($\times 10^{-3}$)	δ ($\times 10^{15}$ cm^{-2})
ZnO/SnO_2 = 50/05	As-dep.	33.86	(110)	352	10.79	11.03	8.59
	Ann. 400 °C	34.71	(311)	339	11.18	10.39	8.01
	Ann. 500 °C	34.52	(311)	369	11.98	9.75	6.97
ZnO/SnO_2 = 50/10	As-dep.	33.82	(110)	150	10.25	11.62	9.50
	Ann. 400 °C	34.56	(311)	195	10.81	10.79	8.55
	Ann. 500 °C	34.56	(311)	233	11.56	10.09	7.48

structure [25] and it could be crystalline if only Zn/Sn ratios become close to 2.0, at a temperature above 500°C [26]. Alternatively, Rajachidambaram *et al.* [27] found that the ZTO films retain its amorphous nature even it is annealed at 600 °C. Particularly, the integration of excess elements, such as Sn, In, Ga, or Al, etc. into the ZnO matrix tempts the alteration into the amorphous form instead of the crystalline phase [28], [29]. Thus, the ZTO films prepared in low RF power of SnO_2 which certainly have low Sn concentration are showing higher peak intensity than the high RF power as seen in the peak intensity values (Table 1). The crystallite size (D) of the films was estimated using the well-known Scherer's formula [30].

The poor crystalline nature as seen in Figure 2 implies that films belong to a higher number of lattice misfit and lattice strain. It is known that the lattice strains could be developed in the films via scattered grains distribution and/or relocation of the atoms from their reference-lattice positions. However, these phenomena are in turn depends on the films' predation conditions including deposition parameters and sub-sequent annealing conditions. Simply, the lattice strain that developed in the film can be known by estimating ''micro-strain''. Alternatively, the atoms that displace from its reference lattice could act as interstitial atoms and its reference place could act as an unlike vacancy in the crystals. The number of atoms that displaced from their reference lattice could be realized via estimating the dislocation density.

The micro-strain (ε) and dislocation density (δ) that indicate the defectiveness of a crystal associated with mis-registry of the lattices and atoms could be calculated using the following equations [30]:

$$\varepsilon = \frac{\beta}{4\tan\theta} \tag{1}$$

And

$$\delta = \frac{1}{D^2} \tag{2}$$

where θ, β, and D have their usual significances as mentioned in the above section. All the estimated values, such as D, ε, and δ for ZTO thin films are tabulated in Table 1 and compared concerning the RF power and subsequent

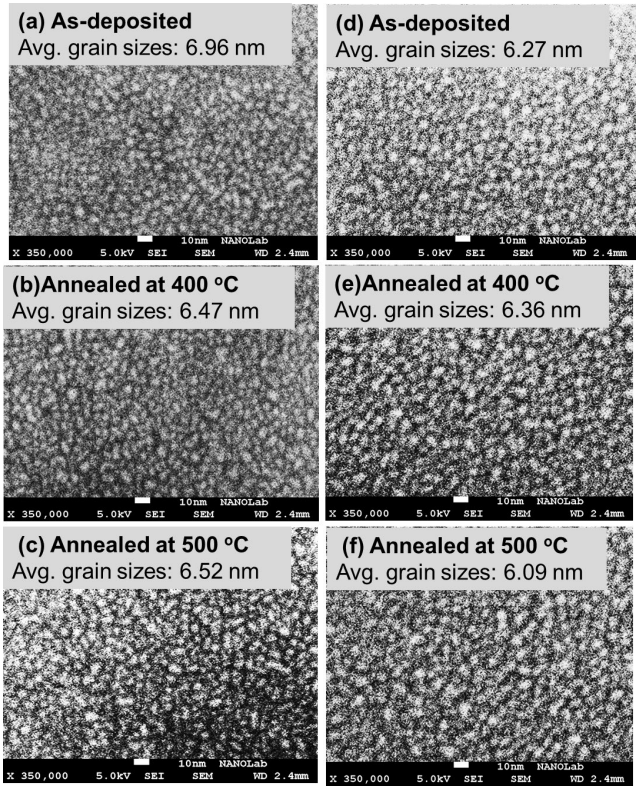

FIGURE 3. FESEM images of ZTO thin films deposited in Ar ambient and subsequent oxygenation in N_2: O_2 ambient, (a)-(c) for RF power of ZnO/ SnO_2 (50/05W) and (d)-(e) for RF power of ZnO/ SnO_2 (50/10W) (average grain size has been estimated by ImageJ software).

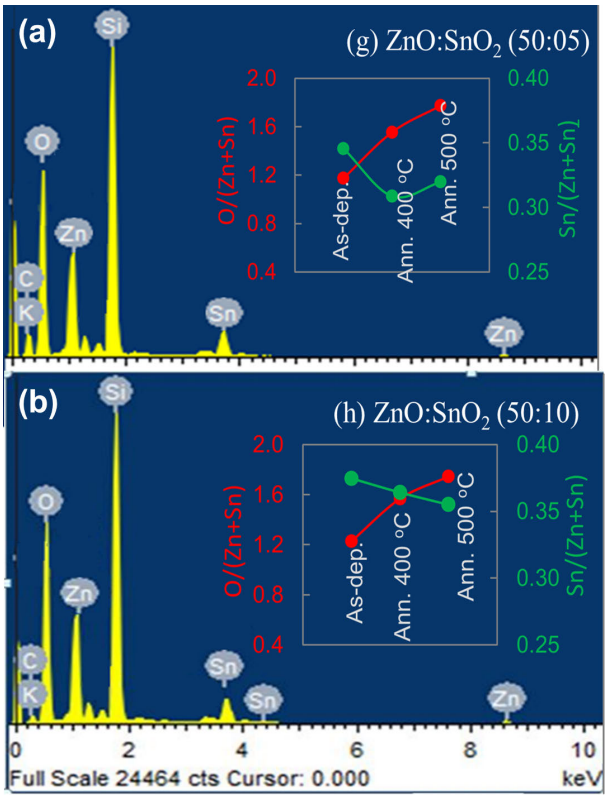

FIGURE 4. (a) and (b) EDX spectra of the film that was at annealed at 500° C in mixed N_2: O_2 ambient (inset figure shows the difference of elemental compositions and the lines are drawn for guiding the eyes).

annealing temperature. It could be seen in Table 1 that the values D of the ZTO films are increased with the increase of annealing time. The micro-strain of the films is found very high, which does not show any significant change by annealing temperature may be due to lack of enough energy supply during annealing that is needed for reconstructing the grains. However, dislocation densities are abridged as the increase of annealing temperature suggests that the interstitial defects which lead to lattice misfit and/or dislocation are reduced in the film by annealing. Overall, the lowest micro-strain, which is the indicator of lattice misfits, is found for 5 watts of SnO_2 film after annealing at 500°C stipulated the strains are released at this temperature comparatively higher than other films.

B. EVALUATING MORPHOLOGICAL & OPTICAL PROPERTIES OF FILM

Films' surface morphology has been investigated via FESEM images as shown in Figure 3. The surface of the films is found dense with nano-platelets; shows very tinny nano-structured; and spherical shaped grains are well distributed over the entire substrate surface. The film's average grain sizes have been estimated by employing ImageJ software [31] and could be seen in Figure 3 that the grain size is decreased slightly with the increase of SnO_2 deposition power. The average grain size of the films deposited at RF power of ZnO/ SnO_2 (50/05W) is 6.65 nm and the RF power of ZnO/ SnO_2 (50/05W) is 6.24 nm. The compositional analysis as shown in Figure 4(a) and 4(b) were corroborated by the EDX result, which for only the films annealed at 500°C. The EDX spectrum confirmed the presence of Zn, Sn, and O elements in the prepared films. The films' elemental compositions could be seen in the insight of Figure 4(a) and 4(b) presented in ratios of Sn/(Zn+Sn) and O/(Zn+Sn) for all films. It could be seen that the atomic concentration of Sn is reduced where O is increased with the increase of annealing temperature that leads to the variation of the optoelectronic properties discussed in the next section. The initial atomic ratio of O with the sum of Zn and Sn is about to 1.2, which increased about to 1.8 upon thermal oxygenation. As the concentration of O is increased, the film's crystal phase changed from $ZnSnO_3$ to $ZnSnO_4$ as seen in XRD. Besides, the Si, K, and C signals have also appeared from the substrate and the trace elements.

Optical transmittance curves of ZTO films deposited by different RF power and subsequent annealing are shown in Figure 5(a) and 5(b). All the films secured an admirable optical transmittance of above 85% in the range of 400 to 900 nm which is a vital need for using a thin film as an HRT buffer layer in thin-film heterojunction solar cells. Moreover, the transmittance increases in the shorter wavelength region (blue shift) is seen for all the annealed films that may be related to the oxygen incorporation into the films.

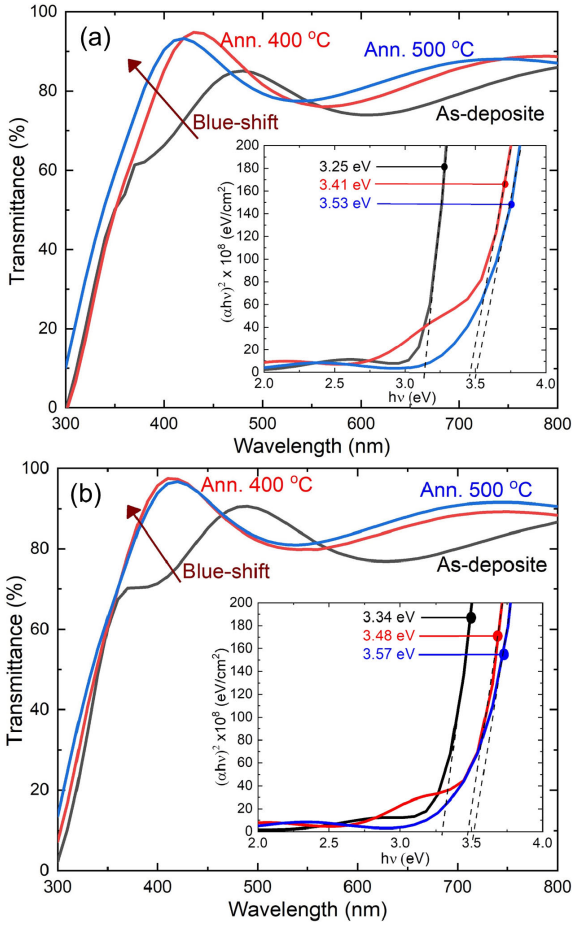

FIGURE 5. Transmittance curves of co-sputtered and annealed ZTO thin films, (a) for RF power of ZnO: SnO_2 (50/05W) and (b) for RF power of ZnO: SnO_2 (50/10W) (inset: bandgap evaluation curves for the corresponding films).

Inset of Figures 5(a) and 5(b) depict the hν vs. $(\alpha h\nu)^2$ plots enabling the E_g to be determined [32]. Comparing with the EDX composition represent in the previous section, it could be realized that the bandgap is increased by the decrease of Sn concentration as well as by the increase of O concentration availed during the post-deposition thermal annealing. It is well known that the bandgap is oppositely changed with the grain size in the case of thin films, this study also shows similar results as compared to SEM images with the films' bandgap. In this case, the elemental composition certainly played a domineering role. The variation of the bandgap may also be corresponding to the film's crystallographic variations as seen in Figure 2. The bandgap found in this study is ranging from 3.25 eV to 3.57 eV which is consistent with the previous study published elsewhere [22], [33].

C. INVESTIGATION ON ELECTRICAL PROPERTIES

The electronic properties of the films were investigated by employing the Hall-Effects measurement system as shown in Table 2. It could be seen that the resistivity of the films increased with the increase of annealing or oxygenation temperature. Alternatively, the resistivity increased with the

TABLE 2. Resistivity, mobility, and carrier concentration of ZTO thin films deposited via different RF power of ZnO and SnO_2 and subsequent oxygenation via thermal annealing in N_2: O_2 ambient.

RF power (ZnO/SnO_2)	Film condition	Resistivity (Ω-cm)	Mobility (cm^2/V-S)	Carrier conc. ($\times 10^{16}$ cm^{-3})
50W/05W	As-dep.	4.32	16.5	8.5
	400°C	21.2	17.9	1.7
	500°C	49.1	10.8	1.2
50W/10W	As-dep.	2.06	17.5	15.1
	400°C	4.42	19.7	7.9
	500°C	12.2	14.3	3.8

increase of O and Zn atomic concentration. The highest resistivity of 48.1 Ω-cm is seen for the annealed (500°C) film of RF power 50/05W and the minimum resistivity of 2.06 Ω-cm is seen for the as-deposited film of RF power 50/10W. This radical change of resistivity is suspected due to the increase of O atoms and reduction of Sn atoms in the film as could be seen in Figure 4(a) and 4(b). However, it is seen that the film's resistivity is directly correlated (inversely varied) with the change of carrier concentration. It was reported that the relatively low carrier concentrations and/or high resistivity are linked to the carrier conduction led by grain boundary scattering that developed by enrichment of oxygen atoms into the films and/or oxygen adsorption onto the grain-boundary of the films [34], [35]. In general, the carrier concentration could be improved by the oxygen-vacancy creation in ZTO thin films via annealing under an oxygen-free ambient [36], however, it is difficult to precisely control the carrier concentrations by oxygen reduction process because it is highly sensitive to process conditions. Besides, carrier concentration could also be improved by the increase of Sn concentration, since valence electrons of Sn and Zn are four and two, thus, Sn could donate two extra electrons when it substituting the Zn atoms or occupying an interstitial site [36].

In the case of semiconductors thin films, carrier concentration, and mobility are considered two key factors that control the carrier transport properties. The significant impact of RF power and subsequent annealing temperature on carrier mobility has been observed. The best mobility as high as 20–50 cm^2/Vs have been reported so far for the film deposited by sputtering and post-annealed at 600°C [25]. In this study, carrier mobility is found to be in the range of 10.8 cm^2/Vs to 19.7 cm^2/Vs and the utmost mobility is attained for the film of RF power 50/10W after 400°C of thermal annealing in an oxygen ambient. It should be noted that the mobility is somehow affected by the deposition condition as it was reported for ZnO thin films grown by sputtering technique [37]. However, the variation of the mobility due to the thermal oxygenation indicating the level of defect density in the films. As the films of the highest O concentration show low mobility, thus, it is reasonably possible that grain boundary scattering is dominated in the carrier conduction in this case as we mentioned earlier. Also, the simultaneous reduction of carrier concentration and mobility in some films suggesting the ionized defect density including sub-bandgap defect density might increase as an impact of thermal annealing [38].

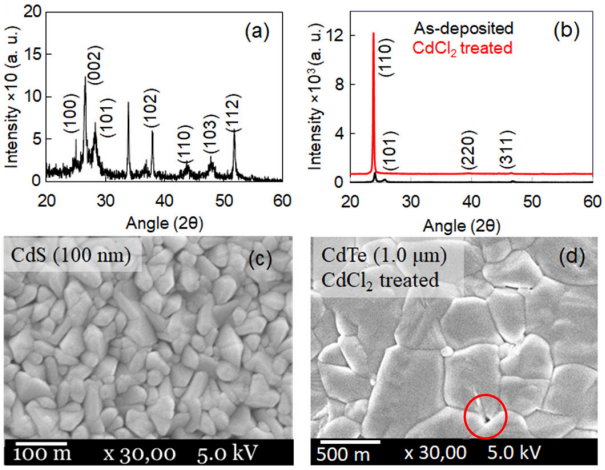

FIGURE 6. XRD spectrum of (a) CdS thin film and (b) CdTe thin-film XRD and FESEM image of (c) CdS and (b) CdTe thin film (marked red circle show the pinhole in the CdTe thin film).

The above analysis clarifies that the film composition and subsequent annealing have a crucial role in film electronic properties, certainly, that brought a great impact on the performance of thin-film solar cells.

D. EVALUATION ON SOLAR CELL PERFORMANCE

Complete solar cells have been primarily fabricated for investigating the effect of ZTO film as an HRT on solar cell performance. For completing the solar cell, the CdS and CdTe thin films have been prepared by sputtering technique at a growth rate of 1.8 Å/s, and 4.6 Å/s, respectively on top of "glass/FTO/ZTO" stacks. Both films were deposited using a substrate temperature of 300 °C and a working pressure of 14 mT. The film thickness of CdS is about to 100 nm and CdTe is about to 2.0 μm. The crystallographic and surface properties of the CdS and CdTe thin films including $CdCl_2$ treated CdTe thin film are shown by XRD and SEM images (Figure 6). XRD spectra show poly-crystalline CdS and CdTe film with preferential orientation (220) and (110), respectively. The crystallinity of CdTe has significantly changed after the $CdCl_2$ treatment, which has been performed by dipping the FTO/ZTO/CdS/CdTe stacks on to the 0.3 M of $CdCl_2$ solution and subsequent vacuum annealing at 390°C for 15 min. Readers are referred to our previous publication regarding the details of the $CdCl_2$ treatment [2]. The inhomogeneous grains with an average grain size of 100 nm are mainly observed in the FESEM image of CdS. The uniformly distributed grains are covered the whole surface homogenously and no pinholes and/or cracks are observed. Instead, the average grain CdTe thin-film after treated by $CdCl_2$ is about to 700 nm, however, some tiny holes are observed as marked in the FESEM image that may affect the cell performance.

The schematic structure of FTO/ZTO/CdS/CdTe/C: Cu/Ag solar cell is shown in the inset of Figure 7(a). The light J-V characteristics of the solar cells are shown in Figure 7(a) and the performance parameters of individual solar cells are shown in Table 3. Diode ideality factor (n), saturation current

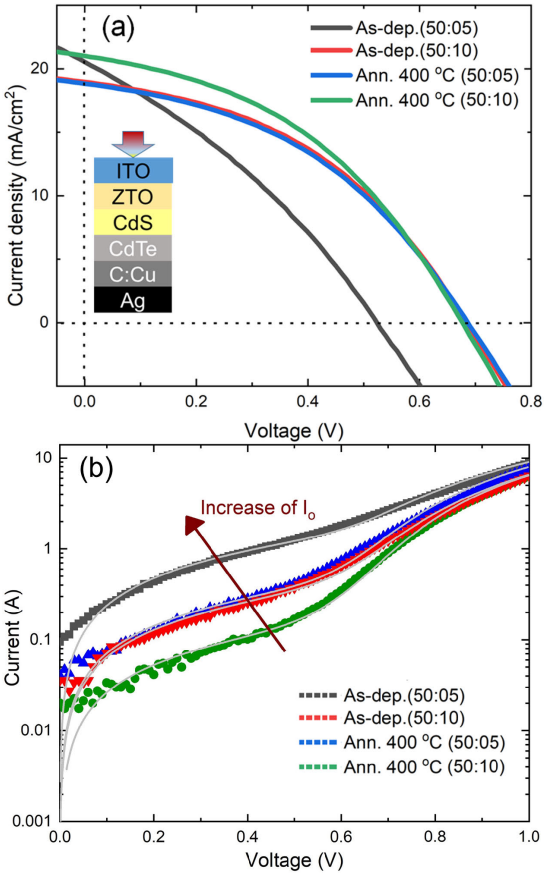

FIGURE 7. (a) Light J-V curve and (b) dark I-V curves in the semi-log mode for FTO/ZTO/CdS/CdTe/C: Cu/Ag solar cells.

TABLE 3. Characteristic parameters of the CdS/CdTe solar cells using ZTO as an HRT buffer layer and C: Cu/Ag printed back contact.

Deposition condition	Jsc (mA/cm²)	Voc (V)	FF (%)	η (%)	I_o (×10⁻⁹, mA/cm²)	n	Rs (Ω-cm²)	Rsh (Ω-cm²)
As-dep. (50/05)	20.54	0.53	35	3.81	13.36	2.39	321	152
As-dep.(50/10)	19.79	0.69	46	6.28	3.91	2.18	314	238
Ann. 400 °C (50/05)	19.83	0.69	46	6.29	5.08	2.25	336	318
Ann. 400 °C (50/10)	21.02	0.67	49	6.90	1.97	1.83	291	378

density (Io), shunt (R_{sh}), and series resistance (R_s) have been extracted by the fitting of dark I-V curves as shown in Figure 7(b) using the following diode equation [39].

$$I(V) = I_o\left[exp\left(\frac{q(V - IR_s)}{nkT}\right) - 1\right] + \frac{V - IR_s}{R_{sh}} \quad (3)$$

where, q is the elementary charge, k the Boltzmann constant, and T the absolute temperature (300 K). I_o, R_{sh}, and R_s were positive, and n was limited to greater than one. The diode fitting was performed using the OriginPro2015J. The estimated parameters are also shown in Table 3.

It could be seen that all the solar cells showed low open-circuit voltage (V_{oc}) and fill factor (FF), which may be attributed to the low Rsh that was found in the dark I-V analysis as shown in Table 3. It has been seen in SEM image that CdTe films have very tiny pin-hole which may contribute to the low shunt resistance. Other causes of low shunt resistance

may be related to the Cu migration through the CdTe grain boundaries to the depletion region which is considered a very critical issue for fabricating ultra-thin (≈2.0 μm) CdTe solar cells. Particularly, higher efficiency in CdTe based solar cells achieved for CdTe thickness of above 5.0 μm [31]. Although the R_{sh} has improved for the annealed ZTO films, however, the cell FF, as well as efficiencies, were not found in the adequate range, owing to the high R_s may be associated with the ZTO HRT layer at the front contact. The thickness of the ZTO film is around 100 nm in this study, we believe that reducing the ZTO thickness certainly improve cell performance. Moreover, the bulk resistance of the CdS and CdTe layers and back contact resistance could also contribute to the increase of series resistance. The high saturation current density (I_o) has also been observed for all the solar cells indicates significant recombination occurred via defect levels where a large number of defects existed within the depletion region that increase due to the Cu ions migration. The high defect density in the depletion region is a cause for low V_{oc}.

The highest value of I_o is 13.36 x10^{-9} mA/cm^2 has been found for as-deposited ZTO film of RF power of 50/05 W. Alternatively, the lowest Io has observed for the ZTO film of RF power 50/10W that was annealed at 400°C. It should be noted that I_o in a solar cell is highly dependent on the generation and recombination of charge carriers in the space-charge region [39]. Particularly, decreasing the structural defect density could lead to a decrease of defect density, thus decreasing the recombination current in the depletion region and I_o in the device [40]. In the case of CdTe devices, the device properties are dominated by recombination at the CdS/CdTe interface which in general has higher defect density due to the large lattice mismatch. However, it could be observed from Table 3 that the ZTO/CdS interface played an important role along with the CdS/CdTe interface which influences the device properties including I_o. Thus, the lowest Io has been observed for the film that has the best crystal properties, highest mobility, and/or lowest defect density. Moreover, the ZTO buffer layer may influence the properties of CdS emitter and CdTe absorber films, as it has been reported that the ZTO buffer layer is promoted to increase the grain size of perovskite films [41].

It is evident that higher efficiency could be achieved by employing the ZTO HRT buffer layer but essential to optimize the FTO/ZTO bilayer properties in the front contact as well as C: Cu/Ag screen printed back contact to create effective ohmic contact for the CdTe solar cells. As a whole, the maximum conversion efficiency of 6.90% has been achieved for the ZTO film of RF power 50/10W that annealed at 400°C including the performance parameters of Jsc of 21.02 mA/cm^2, Voc of 0.67 V, and FF of 49%.

IV. CONCLUSION

We demonstrated extensively the impact of elemental composition on properties of Zin-Tin-Oxide (ZTO) thin films that have been tailored by RF power in the co-sputtering technique and subsequent thermal oxygenation. The oxygenated films showed better crystallinity with a phase transformation from Zn_2SnO_3 to Zn_2SnO_4, however, there has no significant effect been observed in the mean crystallite size, dislocation density, and micro-strain for as-deposited and oxygenated films. All the films showed uniform nanostructure with well-distributed grains that reduce by an increase of SnO_2's deposition power as well as oxygenation temperature. The optical transmission of the films has been found higher than 85% in the visible region and blue-shift is observed in the oxygenated films. The bandgap increases with the increase of O concentration and oxygenation temperature as well. The significant effect of Sn and O concentration on electronic properties has been observed whereby the carrier mobility was found as high as 19.7 cm^2/Vs. The electronic properties of the films are seemed to be affected by the grain boundary scattering that is developed by enrichment of oxygen atoms into the films and/or oxygen adsorption onto the grain-boundary of the films due to the oxygenation. In this study, the optimum optoelectronic properties found for RF power of 50W (ZnO) and 10W (SnO_2) via thermal oxygenation at 400°C where the ratio O/(Zn+Sn) become around 1.6. The complete solar cells with a novel structure of FTO/ZTO/CdS/CdTe/C: Cu/Ag have been fabricated and as high as 6.9% conversion efficiency was achieved so far. Although, the cells are suffered from the low shunt and high series resistance including very high recombination current, however, the significant effect of oxygenation has already been realized as the cell with oxygenated film ($ZnSnO_4$) showed higher efficiency than the cell with as-deposited ($ZnSnO_3$) film. The findings urge the importance of the optimization of ZTO thin films including CdS and CdTe layers for achieving higher efficiency in the ultra-thin solar cell.

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