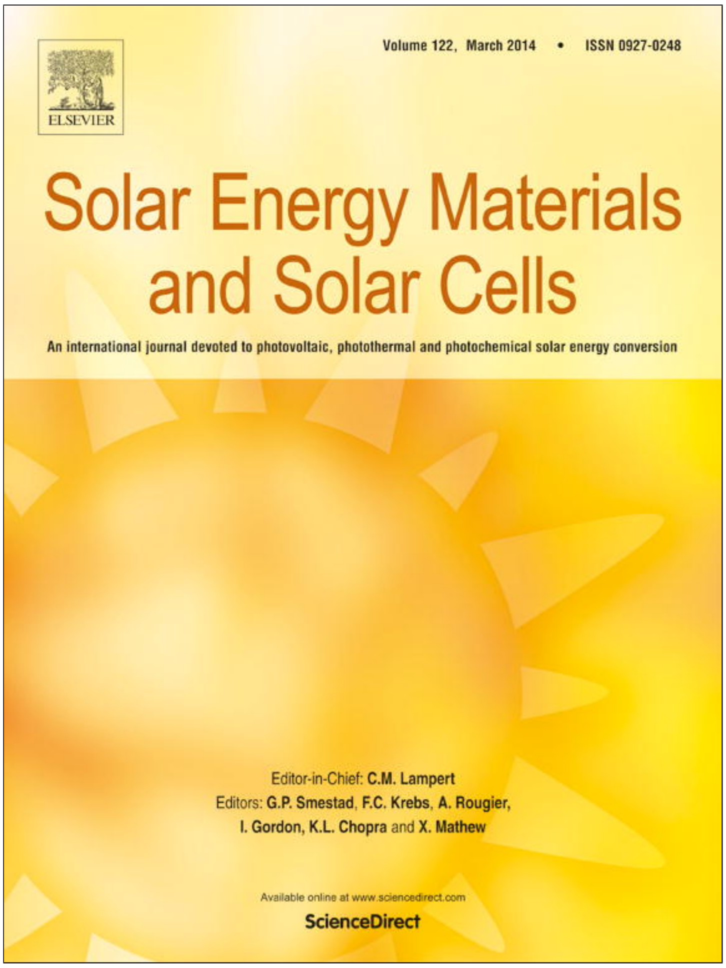
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Influence of high work function ITO:Zr films for the barrier height modification in a-Si:H/c-Si heterojunction solar cells

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ABSTRACT

We report the influence of magnetron sputtered zirconium-doped indium tin oxide (ITO:Zr) films with high mobility and work function on the heterojunction with intrinsic thin layer (HIT) solar cell. The addition of oxygen (O_2) to argon (Ar) flow ratio during the deposition process improves the Hall mobility of the ITO:Zr films while the carrier concentration decreased. The small amount of oxygen resulted in an enhancement of work function while excess amount of O_2 was not suitable for the electrical and surface properties of ITO:Zr films. The increase of O_2/Ar flow ratio from 0% to 0.4% improved the work function from 5.03 to 5.13 eV while the conductivity of ITO:Zr films remained about the same. The ITO:Zr films were employed as a front anti-reflection layer in a HIT solar cell and the best photo-voltage parameters were found to be V_{oc}=710 mV, J_{sc}=33.66 mA/cm^2, FF=72.4%, and η=17.31% for the O_2/Ar flow ratio of 0.4%. The increase of ITO:Zr work function leads to an increase in open circuit voltage (V_{oc}) and fill factor (FF) of the device. Therefore, the ITO:Zr films with high work function can be used to modify the front barrier height in the HIT solar cell.

1. Introduction

The HIT solar cell is considered as a unique photo-voltaic device which resulted from the amorphous silicon (a-Si) and crystalline silicon (c-Si) solar cell technologies. It offers low-cost fabrication and better temperature coefficient as compared to crystalline silicon solar cells with diffused p–n junctions. The performance of HIT solar cell is limited by several factors. The work function of transparent conductive oxide (Φ_{TCO}) is one of the factors which produces the band bending effect in TCO/a-Si:H(p) interface of the HIT solar cell [1]. Due to the difference in the work function of TCO and a-Si:H(p) (Φ_{TCO} and $\Phi_{a\text{-}Si:H(p)}$) layers, an electron injection barrier is developed which limits the hole carriers flow from a-Si:H(p) to TCO layer. The TCO films with high work function are used to inject holes carriers in front contact barrier height of HIT solar cells; hence as high as possible values of work function are desired [2,3].

The indium tin oxide (ITO) films are commonly used as anti-reflection electrodes due to their high conductivity, transmittance in visible wavelength region and wide optical bandgap for the flat panel displays, organic light emitting devices (OLEDs) and photo-voltaic devices [4,5]. However, certain applications like large area flat panel displays and heterojunction solar cells require further improvement in optical and surface properties of the ITO films. Recently, TCO films with low carrier concentration, high mobility and work function are proposed beneficial as front electrode in the HIT solar cell due to free-carrier absorption in the NIR wavelength region and low Schottky barrier height in the front TCO/a-Si:H(p) interface. Zhang et al. suggested that ITO films doped with a small amount of ZrO_2 not only retain the basic characteristics of ITO films but also improve some of the properties like NIR transmittance, chemical and thermal stability of films [6,7,13,14,16]. The ITO films doped with a small amount of high permittivity materials like ZrO_2 resulted in an enhancement of the Hall mobility and NIR transmittance as observed by Gessert et al. [8,21]. Recently, Hussain et al. showed the superior electrical and optical characteristics of ITO:Zr as compared to ITO flms by RF magnetron sputtering for HIT solar cells [22]. Therefore, the ITO:Zr films with high mobility and work function are proposed for the future solar cell applications due to their excellent surface and optical properties [6–8].

Several methods have been adopted to change the ITO work function (Φ_{ITO}) such as by the variation of substrate temperature (T_s) and oxygen (O_2) flow rate [2,10]. The simulation studies have been performed by Rached [3], Zhao [11] and Centurioni et al. [12]

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for the precise role of Φ_{ITO} on the performance of the HIT solar cell. Even though several studies are available related to the electrical and optical properties of ITO:Zr films, however up to our knowledge the use of high mobility and work function ITO:Zr films for HIT solar cells by the variation of O_2/Ar flow ratio is yet to be reported.

In this article, we report the influence of high mobility and work function of ITO:Zr films on the performance of HIT solar cell by the variation of O_2/Ar flow ratio. The electrical, optical and surface properties of the ITO:Zr films are described as a function of O_2/Ar flow ratio. The performance of HIT solar cell for the process parameters like film thickness and O_2/Ar flow ratio is explained with the help of ITO:Zr work function. The precise role of ITO:Zr work function on the band diagram of HIT solar cell is also discussed.

2. Experimental details

The ITO:Zr films were deposited on (1.5 × 1.5 cm^2) the Corning glass (Eagle, 2000) substrates by RF magnetron sputtering system. The ITO:Zr sputter target was composed of $In_2O_3/SnO_2/ZrO_2$ at 90/9.8/0.2 wt% with 99.999% purity. The base pressure of the chamber was maintained as 9×10^{-5} Torr while the working pressure was fixed as 1.5×10^{-3} Torr. The substrate temperature was kept constant at 200 °C while the O_2/Ar flow ratio was varied from 0% to 1% during the sputtering process. The schematic diagram of the HIT solar cell is shown in Fig. 1. The n-type Czochralski-grown mono-crystalline Si wafers with (1–10 Ω cm and 525 μm) were employed for the preparation of HIT solar cells. The Si wafers experienced standard RCA 1 (H_2O_2–NH_4OH–H_2O) and RCA 2 (H_2O_2–HCl–H_2O) cleaning processes after an ultrasonic treatment. The RF powered plasma-enhanced chemical vapor deposition (PECVD) system was used with the conditions of SiH_4:H_2:B_2H_6 (1%)=3:22:0.09 gas ratios for 3% doping concentration, 14 mW cm^{-2} power to reduce plasma damages at 200 °C substrate temperature and 100 mTorr deposition pressure. The hydrogenated amorphous silicon a-Si:H(p/i) layers were deposited on the front side of c-Si wafer by the PECVD system with the thickness of 7 and 5 nm. On the rear side of c-Si wafer, the a-Si:H(i/n) layers were deposited by the PECVD system with the thickness of 5 and 7 nm. The 80 nm ITO:Zr films were deposited on the front side of the HIT solar cell in the presence of a metal mask that was directly placed on the a-Si:H(p) coated surface to form a square-shaped ITO:Zr layer. Finally, the Ag/Al and Al electrodes (~1000 Å) were deposited on the front and rear surfaces by thermal evaporation for good ohmic contacts. The active area of the HIT solar cell was fixed as 1 × 1 cm^2.

Spectroscopic ellipsometry (Nano-view, SE MF-1000) was used to measure the thickness of the RF magnetron sputtered ITO:Zr films. The electrical properties like carrier concentration (n), resistivity (ρ) and the Hall mobility (μ) of ITO:Zr films were characterized by the Hall effect measurement (Ecopia HMS-3000) system. The optical (transmittance and reflectance) properties of the ITO:Zr films were characterized by the solar cell spectral response/QE/IPCE measurement (QEX7) system at room temperature. The surface morphology and roughness of ITO:Zr films were measured by a high resolution atomic force microscopic (HR-AFM) (SII SPA-300HV) system. The X-ray photoelectron spectroscopic (XPS) system was used with the monochromatic Al K_α (1486.6 eV) source to measure the work function of ITO:Zr films. The performance of HIT solar cells was characterized at room temperature by the current density–voltage (LIV) system under AM 1.5 light illumination. The equilibrium band diagram of front contact barrier height of HIT solar cell was studied by simulations.

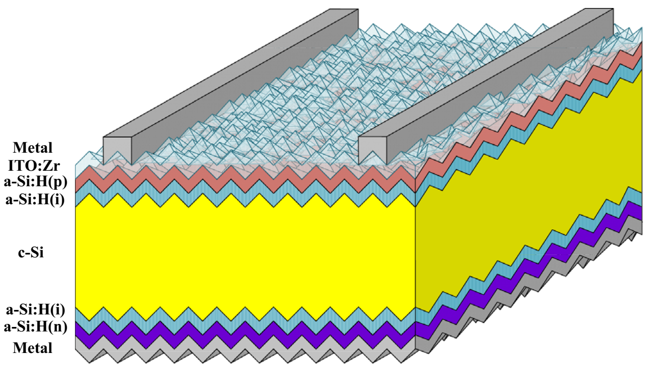

Fig. 1. The schematic diagram of the heterojunction with intrinsic thin layer (HIT) solar cell.

3. Results and discussion

Fig. 2 illustrates the electrical characteristics of ITO:Zr films for various O_2/Ar flow ratios. The resistivity of ITO:Zr films was increased from 3.53×10^{-4} to 4.39×10^{-4} Ω cm with the increase of O_2/Ar flow ratio from 0% to 1%. The slight increase in the resistivity was related to additional oxygen flow during the sputtering process. The combined effect of the Hall mobility and carrier concentration can also be attributed to the resistivity of ITO:Zr films. The sheet resistance of ITO:Zr films was increased from 48.71 to 60.39 Ω/□ with the increase of O_2/Ar flow ratio [13,14]. The Hall mobility of ITO:Zr films improved from 36.2 to 49.2 cm^2/Vs while the carrier concentration decreased from 4.89×10^{20} to 2.89×10^{20} cm^{-3} with the increase of O_2/Ar flow ratio from 0% to 1%. The ITO:Zr films with higher Hall mobility are

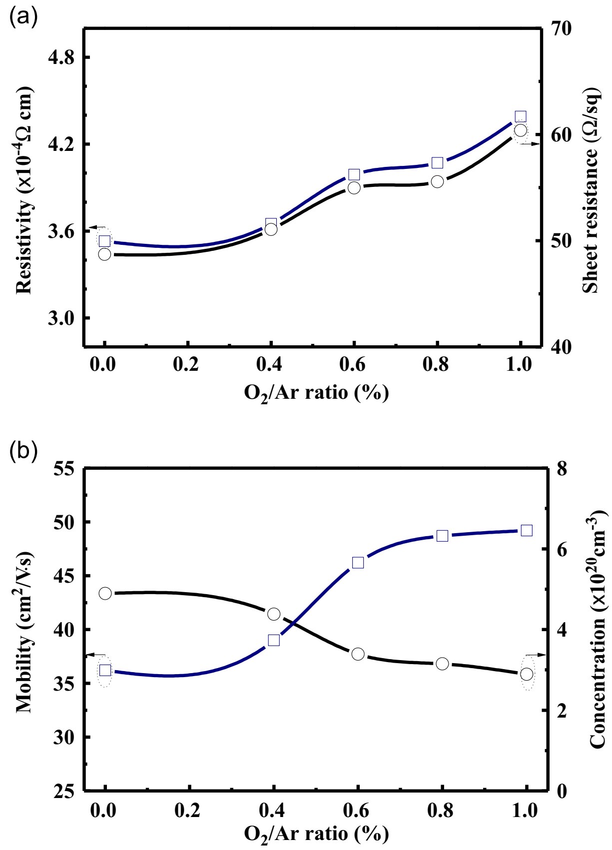

Fig. 2. The electrical characteristics (a) resistivity, sheet resistance and (b) the Hall mobility and carrier concentration of ITO:Zr films for various O_2/Ar flow ratios.

beneficial for HIT solar cells due to lower free-electron concentration. The Hall mobility of ITO:Zr films is dependent on various electron scattering mechanisms i.e. ionized impurity, neutral defect and grain boundary scattering [15]. The decrease in carrier concentration of ITO:Zr films with O_2/Ar flow ratio was due to filling of oxygen vacancies and deactivation of the donor impurities.

The optical transmittance and reflectance spectra of ITO:Zr films for various O_2/Ar flow ratios are shown in Fig. 3(a). The transmittance in the visible wavelength region was increased with the increase of O_2/Ar flow ratio. The as deposited ITO:Zr films showed the transmittance of 85.4% in the visible (400–800 nm) wavelength region. The visible transmittance was further increased from 85.1% to 89.3% with the increase of O_2/Ar flow ratio from 0.4% to 1%. All the ITO:Zr films showed an apparent increase of transmittance in the near-infra-red (NIR) wavelength region as shown by Zhang [6,7] and Gessert et al. [8]. The as deposited ITO:Zr films showed the reflectance of 13.9% in the visible wavelength region that reduced from 13.2% to 12% with the increase of O_2/Ar flow ratio from 0.4% to 0.8%. The reflectance of ITO:Zr films was closely related to the behavior of carrier concentration of films [19,20]. The optical bandgap of the films was obtained from the plot of photon energy (E) versus $(\alpha h\upsilon)^2$ as per the direct transition model. The absorption coefficient (α) was determined from the relation $\alpha=(1/d)\ln(1/T)$, where d and T being the thickness and transmittance of the films [7], respectively. The optical bandgap was decreased from 3.47 to 3.44 eV with the increase of O_2/Ar flow ratio from 0 to 0.6 and then it increased to 3.54 eV with the further increase of O_2/Ar flow ratio.

The surface morphology of ITO:Zr films for various O_2/Ar flow ratios is shown in Fig. 4. The surface roughness of ITO:Zr films was

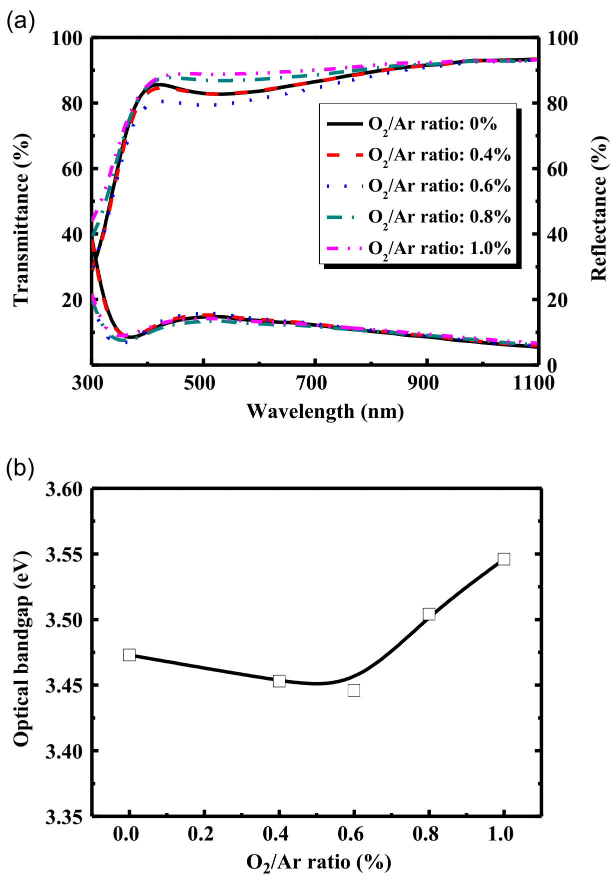

Fig. 3. The optical characteristics (a) transmittance, reflectance and (b) optical bandgap of ITO:Zr films deposited on glass substrates for various O_2/Ar flow ratios.

initially decreased with the increase of O_2/Ar flow ratio from 0% to 0.4%. The ITO:Zr films showed an average roughness (R_a) of 0.4 nm and peak to valley (R_{p-v}) roughness of 19.8 nm for the 0.4% O_2/Ar flow ratio. With the further increase in O_2/Ar flow ratio from 0.6% to 1.0%, the surface roughness of ITO:Zr films rapidly increased due to excess oxygen into the lattices [15,16]. The excessive oxygen can also worsen the surface morphology of the ITO:Zr films. The average roughness of 17.1 nm and peak to valley roughness of 152 nm was recorded for the 1.0% O_2/Ar flow ratio. The surface morphology of 0.4% O_2/Ar flow ratio provided the highest work function and good optical properties. The summary of the AFM parameters of ITO:Zr films is shown in Table 1.

Fig. 5 shows the ITO:Zr work function for various O_2/Ar flow ratios. The work function was estimated from the XPS data and expressed in mathematical form as [17]

$$\Phi_{\mathrm{ITO:Zr}} = h\upsilon - E_B - E_{KE} \quad (1)$$

where $h\upsilon$ is the photon energy, E_B and E_{KE} being the binding energy and kinetic energy of photoelectron, and $\Phi_{\mathrm{ITO:Zr}}$ being the work function of ITO:Zr films. The ITO:Zr work function increased from 5.09 to 5.13 eV with the increase of O_2/Ar flow ratio from 0% to 0.4%. With the further increase of O_2/Ar flow ratio from 0.6% to 1.0%, the work function was decreased from 5.07 to 5.03 eV. The increase of O_2/Ar flow ratio enhanced the $\Phi_{\mathrm{ITO:Zr}}$ which can be attributed to the incorporation of more oxygen onto the surface of ITO:Zr films. As the oxygen concentration on ITO:Zr surface was increased beyond a certain level the work function began to decrease. The oxidative treatment incorporated more oxygen onto ITO surface, and hence the work function can be increased by O_2 addition as observed by Kim. [9] and Mason et al. [18]. The enhancement of $\Phi_{\mathrm{ITO:Zr}}$ attributed to the presence of interfacial dipole caused by the surface rich in negatively charged oxygen. The overall work function of ITO:Zr films was also higher than the already reported work function of ITO films [2,9].

Fig. 6(a) shows the current density–voltage (J–V) characteristics of HIT solar cells for various O_2/Ar flow ratios. The as deposited HIT solar cell showed the short-circuit current density (J_{sc}) of 32.72 mA/cm^2 with an open-circuit voltage (V_{oc}) of 690 mV. An increase of V_{oc} from 690 to 710 mV and J_{sc} of the device from 32.72 to 33.66 mA/cm^2 was observed with the increase of O_2/Ar flow ratio from 0% to 0.4%. The V_{oc} and FF of the device were decreased from 710 to 680 mV and from 72.44% to 70.72% with the further increase of O_2/Ar flow ratio from 0.4% to 1%, respectively. The variation of ITO:Zr work function with O_2/Ar flow ratio influenced the overall efficiency of the HIT solar cell. The change in $\Phi_{\mathrm{ITO:Zr}}$ influenced the FF and V_{oc} of the device as observed by Oh [2], Rached [3], and Hussain et al. [10]. The best photo-voltage parameters of the HIT solar cell were recorded as $V_{oc}=710$ mV, FF=72.4%, $J_{sc}=33.66$ mA/cm^2 and $\eta=17.31\%$ at O_2/Ar flow ratio of 0.4% for $\Phi_{\mathrm{ITO:Zr}}$ of 5.13 eV. Fig. 6(b) shows the J–V characteristics of HIT solar cell as a function of ITO:Zr film thickness. The HIT solar cell showed J_{sc} of 33.66 mA/cm^2 and V_{oc} of 710 mV for ITO:Zr thickness of 80 nm. The J_{sc} and FF of the device were increased from 33.66 to 34.44 mA/cm^2 and from 72.16% to 74.83% with the increase of ITO:Zr film thickness from 80 to 120 nm, respectively. The $\Phi_{\mathrm{ITO:Zr}}$ increased from 5.09 to 5.13 eV with the increase of thickness from 80 to 120 nm. So the increase of J_{sc} and FF improved the overall performance of HIT solar cell. The best photo-voltage parameters were found to be $V_{oc}=710$ mV, $J_{sc}=34.44$ mA/cm^2, FF=74.8%, and $\eta=18.30\%$ for ITO:Zr thickness of 120 nm and work function of 5.13 eV [22]. The variation in V_{oc} and FF was related to $\Phi_{\mathrm{ITO:Zr}}$ while the minor variation in J_{sc} resulted from high mobility and low carrier concentration of ITO:Zr films [10,23]. The ITO:Zr films are proposed as an alternate TCO material for the future high efficiency photovoltaic applications.

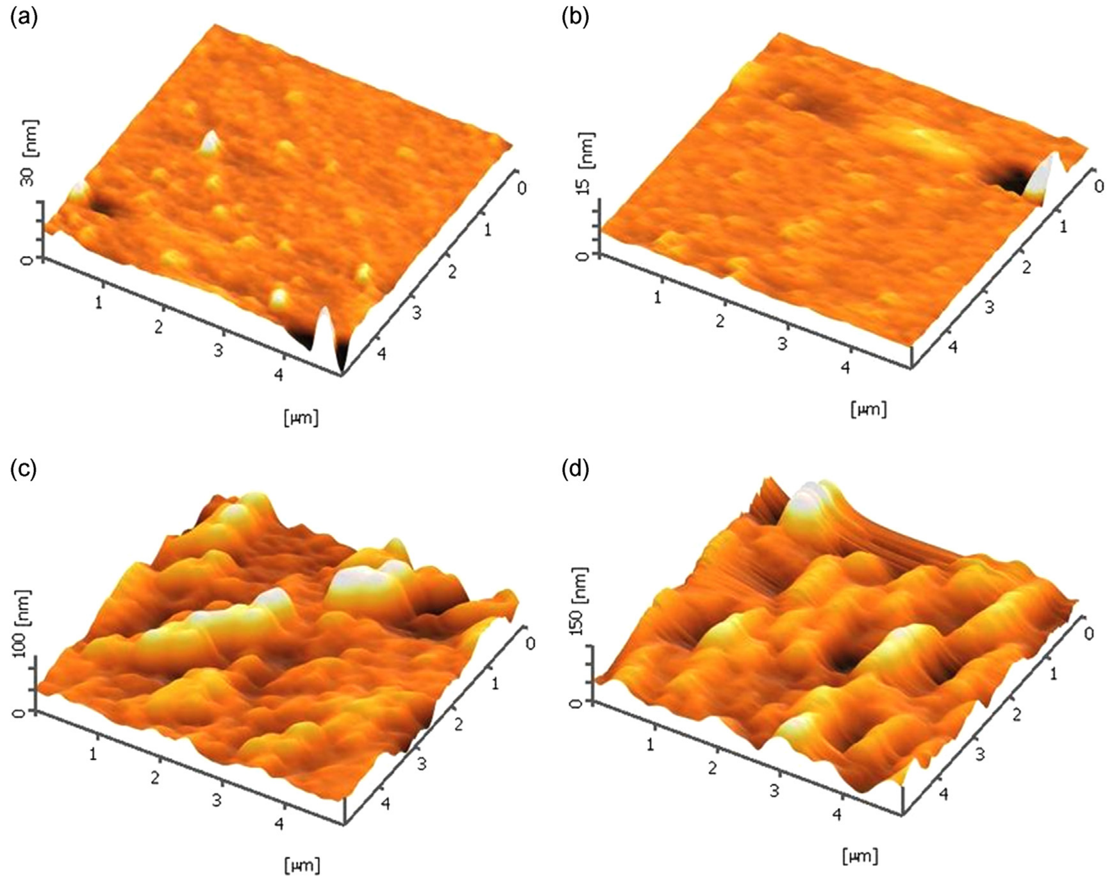

Fig. 4. The AFM images of the ITO:Zr films for the O_2/Ar (a) 0%, (b) 0.4%, (c) 0.8% and (d) 1.0% flow ratios.

Table 1
Summary of AFM measurements for ITO:Zr films as a function of O_2/Ar flow ratio.

O_2/Ar ratio (%)	R_{rms} (nm)	R_a (nm)	R_{p-v} (nm)
0	1.11	0.59	30.87
0.4	0.89	0.40	19.8
0.8	19.3	14.71	131.4
1	21.89	17.1	152

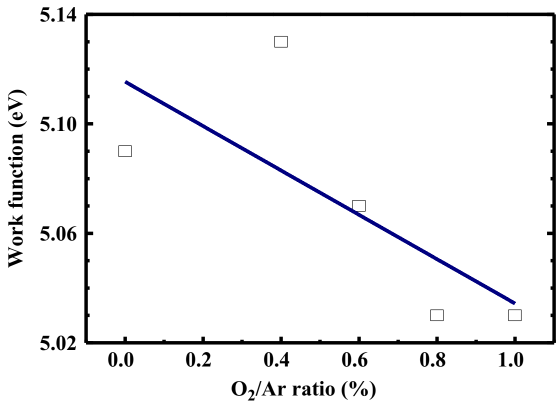

Fig. 5. The work function of ITO:Zr films for various O_2/Ar flow ratios.

Fig. 7 shows the equilibrium band diagram of front contact (ITO:Zr/a-Si:H(p)/a-Si:H(i)/c-Si(n)) barrier height of HIT solar cell as a function of work function. The band bending in the a-Si:H (p) region was resulted from the difference in work function of ITO:Zr and a-Si:H(p) layer and the discrepancy between $\Phi_{a\text{-}Si:H(p)}$ and $\Phi_{c\text{-}Si}$. Due to lower work function of TCO as compared to a-Si:H(p) layer, an electron injection barrier can develop that limits the V_{oc} downward band bending. The enhancement in ITO:Zr work function improved the migration of hole carriers from a-Si:H(p) to ITO:Zr layer and resulted in an increase of FF of HIT solar cells [3,12]. The decrease in electric field at ITO:Zr/a-Si:H(p) interface may be the reason for the reduction of band bending. Similarly with the increase of work function to 5.13 eV, the V_D of ITO:Zr/a-SI:H(p) interface resulted in an increase of built-in voltage and higher V_{oc}. Hence, the overall performance of HIT solar cell was improved with the increase of FF and V_{oc}. As mentioned earlier, the increase in the O_2/Ar flow ratio leads to an enhancement of work function, in turn increase the conduction band discontinuity between c-Si and a-Si:H(p), whereas decreasing the Schottky barrier height between ITO/a-Si:H(p) interface. The increase in conduction band discontinuity prevents the electron diffusing from base to emitter-layer, while decreasing the Schottky barrier height results in an enhancement of hole carrier collection at the front contact.

Fig. 8 depicts the schematic band diagram of the ITO:Zr/a-Si:H (p) interface for HIT solar cell. E_{g1}, E_{g2} and $\Phi_{ITO:Zr}$, $\Phi_{a\text{-}Si:H(p)}$ being the optical bandgaps and work functions of ITO:Zr and a-Si:H (p) layers, respectively. The χ and E_{ac} being the electron affinity and activation energy of a-Si:H(p) layer. Similarly, the Φ_{bo} and E_{sbb} being the front contact barrier height and band bending of ITO:Zr/a-Si:H(p) interface, while V_{bi} and E_b are the build-in potential and the surface potential barrier for ITO:Zr/a-Si:H(p) interface [12], respectively. Due to the wide optical bandgap of ITO:Zr films with degenerated doping (n-type semiconductor), the Fermi level lies in the conduction band. The highly doped ITO:Zr layer behaves

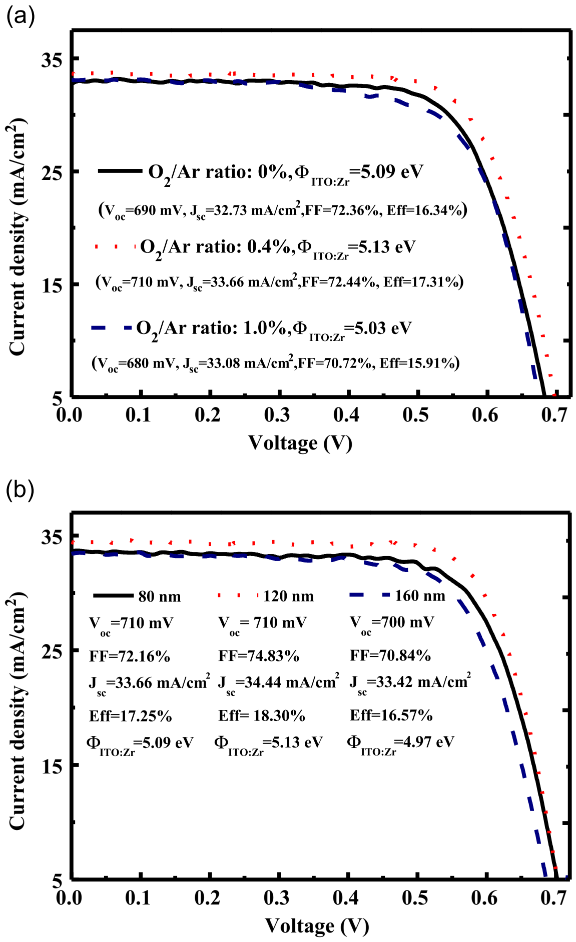

Fig. 6. The (a) current density–voltage (*J–V*) characteristics of the HIT solar cells for various O_2/Ar flow ratios and (b) *J–V* characteristics of HIT solar cells as a function of ITO:Zr film thickness.

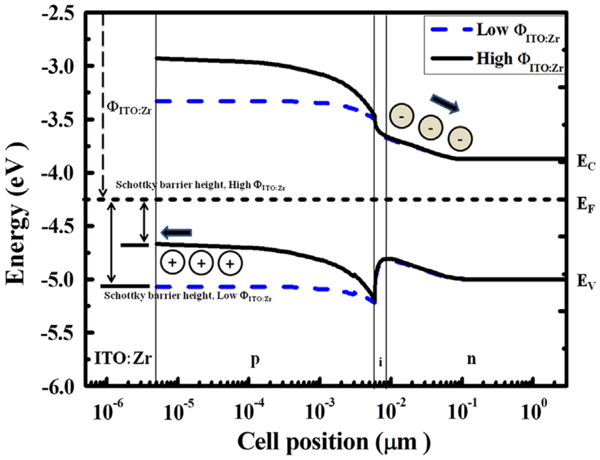

Fig. 7. The band diagram of front contact barrier (ITO:Zr/a-Si:H(p)/a-Si:H(i)/c-Si(n)) height of HIT solar cell as a function of ITO:Zr work function.

electronically like metal and the electronic behavior of the ITO:Zr/*a*-Si:H(p) interface usually assumed as similar to a metal–semiconductor junction [23,24]. The increase in ITO:Zr work function in turn decreased the Schottky barrier height between ITO:Zr and a-Si:H(p) layers, hence more hole carriers can be collected at the front contact.

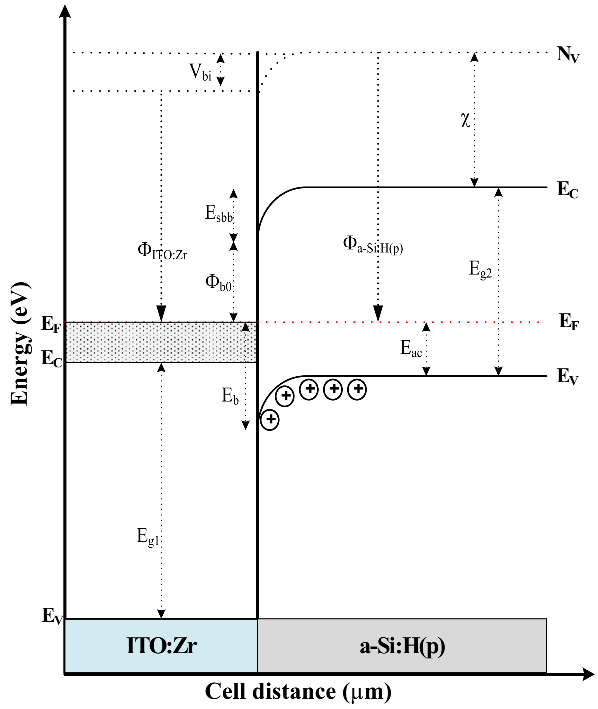

Fig. 8. The ITO:Zr/a-Si:H(p) interface diagram of HIT solar cell as a function of ITO:Zr work function.

4. Conclusion

In summary, we deposited the ITO:Zr films with high work function, mobility and low carrier concentration by RF magnetron sputtering. The surface morphology and work function of the films were improved by low O_2 concentration while higher percentage of O_2 degraded the characteristics of ITO:Zr films. The ITO:Zr work function was derived from XPS measurements and varied from 5.03 to 5.13 eV with the increase of O_2/Ar flow ratio. The ITO:Zr films were employed as a front anti-reflection electrode in HIT solar cells and the best photo-voltage parameters were found to be: $V_{oc}=710$ mV, $J_{sc}=33.66$ mA/cm^2, FF$=0.724$, and $\eta=17.31\%$ for O_2/Ar flow ratio of 0.4% and work function of 5.13 eV. The increase in ITO:Zr work function leads to a band bending effect that improved the V_{oc} and FF of the device. The minor increase in J_{sc} of the HIT solar cell was related to high mobility and low carrier concentration of ITO:Zr films. Therefore, the ITO:Zr films with high mobility and work function are proposed for future high performance photovoltaic applications.

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