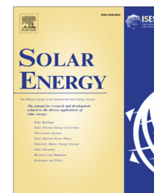
Solar Energy 155 (2017) 963–970

Contents lists available at ScienceDirect

Solar Energy

journal homepage: www.elsevier.com/locate/solener



The bifunctional tin-doped indium oxide as hole-selective contact and collector in silicon heterojunction solar cell with a stable intermediate oxide layer

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ARTICLE INFO

Article history:
Received 21 March 2017
Received in revised form 4 July 2017
Accepted 13 July 2017

Keywords:
Silicon-based heterojunction solar cells
Tin doped indium oxide film
Magnetron sputtering
Work function
Carrier transport

ABSTRACT

The tin-doped indium oxide (ITO) and the intermediate nanometer-scale SiO_x layers were synthesized directly on n-type crystalline silicon (n-Si) substrate by radio-frequency magnetron sputtering deposition. During the ITO-sputtering deposition, the effect of shallow implantation intermixing led to forming an ultra-thin SiO_x layer, which could successfully lessen the interface states and promote the transportation of carriers. The photovoltaic properties of devices showed the open-circuit voltage (V_{oc}) strongly correlated to the carrier concentration of ITO (n_{ITO}), indicating a hole-selective contact of ITO. An equivalent "p-type Fermi level" (hole as majority carriers) was reasonably employed to interpret the decrease of V_{oc} with the increase of n_{ITO}. The impact of the work function difference between ITO and n-Si on V_{oc} of ITO/SiO_x/n-Si heterojunction cells was tentatively equivalent to the difference of the defined quasi-Fermi levels. Through the modification of surface-reflectance and rear contact, the heterojunction structure solar cells achieved efficiency of 11.50 ± 0.17%. Furthermore, the stability of the devices in conversion efficiency was excellent over a whole year. The temperature coefficient of −0.34%/°C was obtained, which was better than −0.45%/°C of a typical diffused-junction silicon solar cell.

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1. Introduction

A strong and stable built-in field established stably in compositional bulk with photovoltaic (PV) function is essential for inorganic solar cells. Because of the research and development in laboratories, high open-circuit voltage (V_{oc}) of more than 700 mV and the reliable opto-electronic conversion efficiency of more than 25% (Zhao et al., 1999; Taguchi et al., 2014; Masuko et al., 2014) have been implemented. Although the above-mentioned solar cells possess nearly perfect PV characteristics, there is still a big challenge on reduction of the complicated processes in the large scale industrial manufacturing. In recent years, the tunnel oxide passivated contact (TOPCon) (Feldmann et al., 2014) was proposed to solve the incompatibility of tunneling and passivating of an oxide layer and has been proved to be feasible (Moldovan et al., 2015; Liu et al., 2016). Another means to facilitating fabrication of solar cells is using dopant-free materials, such as MoO_x (Battaglia et al., 2014; Bivour et al., 2015; Geissbühler et al., 2015; Bullock et al., 2016), V_2O_x (Gerling et al., 2016) and TiO_x (Bullock et al., 2016; Yang et al., 2016), as hole or electron selective contacts to effectively separate electron-hole pairs (Würfel et al., 2015). In fact, the second means is originated from conductor-insulator-semicon ductor (CIS) solar cells (Singh et al., 1981) or semiconductor-insula tor-semiconductor (SIS) solar cells (Kobayashi et al., 1995, 1991; Shewchun et al., 1979; Shewchun et al., 1978; Ghosh et al., 1978). Generally, transparent conducting oxides (TCO) materials such as ITO (Malik et al., 2008) and doped-ZnO (Lupan et al., 2009; Bethge et al., 2013; Fang et al., 2013; Pietruszka et al., 2016) are degenerate n-type semiconductor materials with low resistivity and high transmittance in the visible range of the solar spectrum. Thus, they are deposited on absorbable narrow bandgap semiconductors like silicon to form a chemical heterojunction (TCO/SiO_x(a-Si:H)/Si) together with a functional built-in field.

As same as TOPCon and carriers selective contacts, TCO/SiO_x(or a-Si:H)/Si heterojunction system encounters this situation that carriers could be availably separated, transported and collected, which resulted in low conversion efficiency (less than 10% (Malik et al., 2008; Lupan et al., 2009; Bethge et al., 2013; Fang et al., 2013, 2014; Pietruszka et al., 2016; Untila et al., 2016)). One of the main problems is how the ultra-thin SiO_x or intrinsic a-Si:H

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http://dx.doi.org/10.1016/j.solener.2017.07.042
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could hold effective passivation after high energy plasma radiation in deposition process of TCO materials. TCO materials are usually deposited by magnetron sputtering, which yet have been demonstrated to damage the passivation layer and decrease effective minority carrier lifetime (Demaurex et al., 2012). Therefore, relatively mild techniques, such as successive chemical solution deposition (Lupan et al., 2009; Untila et al., 2016), atomic layer deposition (Bethge et al., 2013) and pulsed laser deposition (Fang et al., 2014), were adopted to deposit TCO layers. However, the industry-wide method of magnetron sputtering deposition should not be neglected in fabricating solar cells.

On the other hand, V_{oc}s were quite different in various SIS solar cells. It is mainly ascribed to the uncontrollable interface states which ought to be passivated by an ultra-thin passivating layer. Besides, the deposition condition of TCO (Cesare et al., 2012), conduction band offset between TCO/Si (Pietruszka et al., 2016), and carrier concentration of TCO (Wenas and Riyadi, 2006) also influence on V_{oc}, yet most of viewpoints were unproven.

In previous work (Du et al., 2015), the transport mechanisms of photo-generated carrier and the formation reasons of stable built-in field in ITO/SiO_x/n-Si solar cells (efficiency of 9.27%) were elucidated in detail. However, there is a big progress in improving the efficiency of such devices.

In this work, radio-frequency (RF) magnetron sputtering was used to deposit ITO layer and the subsequent effects of such deposition method were studied as well. Through changing sputtering condition, the relationship between carrier concentration of ITO (n_{ITO}) and V_{oc} of SIS devices was discussed. By modifying the fabrication conditions, the efficiency of SIS devices was significantly improved. Furthermore, the stability of the device was tested under a continuous irradiation of simulated solar source.

2. Experimental in detail

ITO thin film as a TCO functional material with excellent optical and electronic features (high transparency in visible region and low resistivity) was directly deposited on n-type Si (1 0 0) wafers by RF magnetron sputtering (Ray et al., 1983; Hadj Tahar et al., 1998). The ITO target used for sputtering purpose was a disk of sintered ceramic mixtures (with a diameter of 3.0 in.) of 90 wt.% In_2O_3 and 10 wt.% SnO_2 (both of their purity is in 99.99%). Before transferred into the vacuum chamber, solar-grade n-type Si wafers (carrier density: 2.0×10^{15} cm^{-3}, and work function Φ_{Si}: 4.30 eV) were cleaned by RCA processing. In the last cleaning process, the wafers were dipped in hydrofluoric acid (HF) (5.0 vol%) for 2 min to remove any native oxide, and rinsed in de-ionized water (18.2 MΩ cm) then blown dry with nitrogen. Subsequently, the substrates were transferred to the sputtering chamber in nitrogen atmosphere. The background pressure of the deposition vacuum chamber was 2.0×10^{-6} Torr and the distance between the target and the substrate was 6.0 cm. The substrate temperature and sputtering power were adjusted to control the properties of ITO layer at Argon (high-purity Ar, 99.999%) atmosphere pressure of 1.0 Pa. Typically, the substrate temperatures were kept from 150 °C to 450 °C, and the sputtering powers were changed from 70 W to 130 W. 5 groups of ITO films and ITO/n-Si heterojunctions were obtained. Through TU-1901 spectrophotometer and HL5500-PC Hall effects measurements, the UV–VIS absorption spectra and the semiconductor characteristic parameters of the ITO films were tested (Figs. S1 and S2) and primary data were summarized in Table 1. The thickness of ITO films was precisely controlled and individually determined by the ellipsometry (SENTECH, SE400 adv-PV) and alpha-step profilometer (AMBIOS, XP-2), respectively. Afterward, Al (1 μm)/Ag (100 nm) and Al (2 μm) metals were deposited by thermal evaporation in vacuum as the front grids and the backside electrodes, respectively, to form good Ohmic contact. Note that there was no particular silicon oxidation as an anterior process involved for the other purposes.

Photocurrent density–voltage (*J-V*) characteristics of the SIS-structured photovoltaic devices were measured using a solar cell efficiency measurement system (calibrated by NREL standard c-Si solar cells). It is equipped with a Xenon lamp and a Keithley 2400 I-V source meter under illumination of 100 mW/cm^2 (AM1.5 G) in air. The elemental and/or chemical stoichiometric composition within ITO films and the intermediate region of ITO/c-Si were carried out by an ESCALAB 250Xi spectrometer with Al kα (1486.6 eV) source. The layer by layer Argon ion milling was applied to get the depth etching profile. The energy resolution of the X-ray photoelectron spectroscopy (XPS) is approximately 0.45 eV by a calibration of a Ta_2O_5 standard sample. The cross-section morphology of the interface region and the thickness profiling of ITO films were achieved by JEM-2010F HR-TEM under micrometer-sized tests. The samples were cut by 600i Double Beam Focused Ion Beam. The preferential growth and phase structure of ITO films on Si substrate was observed by X-ray diffraction in a MAX2500V+ system with the high power of 10 kW.

3. Results and discussion

3.1. Interfacial properties between ITO layer and Si substrate

Since the ITO layer was directly deposited on HF-treated n-Si substrate by RF magnetron sputtering, it is necessary to investigate the effects of ITO on surface of Si substrate. Fig. 1 shows the cross-section morphology of ITO/n-Si interface through the HR-TEM. The lattice inside the ITO layer and Si substrate are arranged neatly and uniformly with a set of crystalline plane lines. According to the crystal structure simulations (inset atomic models) of ITO and Si substrate by software of VESTA software (Momma and Izumi, 2008), it is demonstrated that the angle between ITO (2 2 2) and Si (1 0 0) is 77.6°. The excellent crystal array indicates that the light-induced carriers could straightly transport from Si substrate to ITO surface and be collected by front grid electrode without grain boundary scattering. Besides, there is an ultra-thin amorphous interface layer with the uneven thickness of approximately 1.3–1.9 nm existing in interlaced zone of ITO and Si lattices. The inset graph in Fig. 1 clearly shows that the interface bilayer is consisted of bilayer labeled as i-#1 and i-#2. The front layer is of amorphous structure and the second layer is of collapsed lattice (light color) which is different from internal structure of c-Si.

In order to verify the component of this interface layer, XPS depth profiling with Ar^+ etching for the determination of elemental or compositional components is employed in the investigation. The experimental results are illustrated in Fig. 2(a). The aligned etching was along the ITO surface, intermediate region of ITO/Si, and Si substrate, with an interval step of 40 s. The image shows an evolution of the elemental In, Sn, O, and Si atoms and compound of SiO_x around the interfacial region over etching time. It manifests that the ratio of In atoms begin to drop off at 950 s, while the ratio of Si atoms are onset to increase slowly. At the etching time of 1200 s, the ratio of Si atoms become dominant, while In atoms almost vanish. It could be judged that the interfacial region is at the etching time range of 950–1200 s. By analyzing the Si *2p* spectra at this etching time range (Fig. S3), SiO_x, including Si_2O, SiO, Si_2O_3 and SiO_2 components, is found in the interfacial region (Himpsel et al., 1988; Keister, 1999; Seah and Spencer, 2003). Furthermore, it is observed in Fig. 2(a) that both SiO_x and In_2O_3 exist at range from 950 s to 1180 s, indicating i-#1 layer (in inset of Fig. 1) contains mixture of SiO_x and In_2O_3 or $In_{2x}Si_yO_{3x+2y}$ (ISO). After In atoms vanish, residual O element and SiO_x are still observed so

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