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Hydrogen dilution on an undoped silicon oxide layer and Its application to amorphous silicon thin-film solar cells

Yu-Hung Chen *, Chen-Cheng Lin, Yung-Tsung Liu, Hsin-Wei Lu, Jun-Chin Liu

Photovoltaic Technology Division, Green Energy & Environment Research Laboratories, Industrial Technology Research Institute, Hsinchu 31040, Taiwan

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ABSTRACT

This paper proposes the use of undoped hydrogenated microcrystalline silicon oxide (μc-SiO_x:H) deposited on an n-μc-Si:H layer of amorphous silicon single-junction superstrate-configuration thin-film solar cells produced using 40 MHz very high frequency plasma-enhanced chemical vapor deposition. We found that undoped μc-SiO_x:H thin film under optimized hydrogen dilution conditions had high crystallinity, high conductivity, a wide optical band gap, and a high refractive index, which are advantageous properties in solar cells. However, deposition at higher hydrogen dilutions degraded the quality and optoelectronic properties of the films, because the morphology of the films changed from microcrystalline to amorphous. These results suggest that the use of an optimized undoped μc-SiO_x:H layer improves a-Si:H thin-film solar cell performance through enhancement of the short-circuit current density J_{sc}. The increased J_{sc} can be attributed to an improved light-trapping capability in the long wavelength range, between 620 and 680 nm, as demonstrated by the external quantum efficiency. This technique also allows optimal conversion efficiency to be achieved. The results demonstrated that hydrogen dilution plays a dominant role in the improvement of film quality and solar cell performance; however, the tradeoff between refractive index and conductivity must be considered.

1. Introduction

Hydrogenated amorphous silicon (a-Si:H) thin-film solar cells have received considerable attention in photovoltaic research because of their potential to produce electricity from solar energy at a low cost. Amorphous silicon is a direct band gap material, which means that less silicon is used in the production of a-Si cells. It is also a type of thin-film technology in which, rather than being expensively processed from monocrystalline, materials are deposited at temperatures below 300 °C. This allows deposition not only on glass, but also on lightweight flexible substrates, such as stainless steel [1,2]. As the volume of photovoltaic manufacturing increases to meet growing market demand, limiting the environmental impact becomes increasingly difficult. The production of a-Si:H does not require the use of heavy metals (e.g., Cd) or rare elements such as In or Te, which assists in reducing environmental impacts during fabrication, and produce abundant material supplies. These advantages make a-Si:H the most reliable and readily available material for use in thin-film solar cells. Research is now focused on the development of high-efficiency a-Si:H photovoltaic technologies, both to increase the market competitiveness of thin-film solar cells and to improve performance-to-price ratios. The use of microcrystalline silicon oxide (μc-SiO_x) layers in silicon-based solar cells has attracted considerable attention in recent years. Compared with conventional wide-band-gap materials, such as amorphous silicon carbide (a-SiC_x), microcrystalline silicon carbide (μc-SiC_x), or silicon nitride (μc-SiN_x), the optical properties of μc-SiO_x can be tuned over a wider range while maintaining high conductivity and a low absorption coefficient [3–6]. Therefore, when p-μc-SiO_x is used as a window layer on the heterojunction p/i interface in the p–i–n structure of a-Si thin-film solar cells, it effectively improves the short-circuit current density (J_{sc}) and contributes to the low absorption coefficients of the wide-gap p-layer, which leads to an increase in light entering the i-layer [7]. When n-μc-SiO_x acts as an effective replacement for integrated n-μc-Si and ITO, the enhancement of the J_{sc} of the μc-Si single-junction solar cell can be attributed to the refractive index difference between silicon film and Ag, which induces optical reflection back into the absorber [8]. When replacing the a-Si:H n-layer by an n-μc-SiO_x:H layer in an a-Si:H single junction solar cell with Ag back reflector, the parasitic absorption both in the n-layer and at the Ag back reflector is reduced, which results in an increase in J_{sc} [23]. When p-μc-SiO:H is used as the window layer in crystalline silicon heterojunction solar cells, the increase in J_{sc} is attributable to the improvement in short-wavelength-region optical transmittance. The higher conductivity of the p-μc-SiO:H window layer

* Corresponding author. Fax: +886 3 5822157.
E-mail address: YHChen@itri.org.tw (Y.-H. Chen).

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leads to low contact resistance between the p-μc-SiO:H and ITO layers. The increase in fill factor (FF) is therefore caused by the improvement in electrical properties [9]. In our recent study [10], we reported that for undoped μc-SiO_x:H thin films, adding a small amount of oxygen into the μc-network results in a low optical absorption, wide band gap, high optical band gap E_{04}, high refractive index, reasonable conductivity, and crystalline volume fraction. On the basis of these findings, undoped μc-SiO_x:H films were deposited on the n-μc-Si:H layer of a-Si:H single-junction superstrate-configuration thin-film solar cells, which improved J_{sc}, primarily because of effective light reflection in the long wavelength region near the i/n interface. Undoped μc-SiO_x:H thin films were proven to enhance the performance of a-Si:H solar cells. Furthermore, the decrease in conductivity was correlated with increased levels of oxygen in the μc-Si:H film. The excessive oxygen content in the μc-SiO_x:H weakened its electrical conductivity, demonstrating that oxygen plays a critical role in the growth of crystallites in SiO:H films. In this study, we investigated the microstructure and optoelectronic properties of undoped μc-SiO_x:H films with varying levels of H_2 dilution. We also fabricated a-Si:H single-junction p–i–n solar cells containing an undoped μc-SiO_x:H layer to evaluate the quality of the undoped μc-SiO_x:H material and cell performance.

2. Experimental

Undoped microcrystalline silicon oxide (μc-SiO_x:H) films were prepared through a 40 MHz very high frequency plasma-enhanced chemical vapor deposition cluster system, using a mixture of silane (SiH_4), hydrogen (H_2), and nitrous oxide (N_2O) gases. The deposition conditions are summarized in Table 1. The structure of the films was studied using Raman spectroscopy (Jobin-Yvon T64000 spectrometer) to measure the crystalline volume fraction. The optical bandgap E_{opt} (the Tauc gap) and the refractive index (n) were estimated from the transmission spectra, which were measured using a Perkin Elmer LAMBDA 750S UV/Vis/NIR spectrophotometer. The electrical properties of the films were examined on the basis of coplanar conductivity, which was measured using an Ag electrode. For p–i–n silicon thin film deposition, we have used multi-chamber cluster 40 MHz PECVD system. In order to prevent cross contamination of impurities, etch layer is prepared in a separate chamber. The optical band gap of the intrinsic a-Si:H thin films was 1.75 eV, as determined using the conventional Tauc plot method. The photosensitivity of the a-Si:H thin films was approximately five orders of magnitude. Boron-doped amorphous silicon oxide (p-a-SiO:H) layers were deposited using $B_2H_6/SiH_4/N_2O/H_2$ gas mixtures, and the p-layer thickness was around 10 nm. Phosphorus-doped microcrystalline silicon n-layers (n-μc-Si:H) were then deposited in a mixed atmosphere of $PH_3/SiH_4/H_2$ gases to a thickness of 9 nm. A 50 nm zinc oxide (ZnO) and 200 nm Ag were subsequently deposited on the n-layer by pulsed DC magnetron sputtering as the back TCO and metal contact layers. A detailed description of the fabrication process has been reported elsewhere [10–11,15–17]. A schematic view of the a-Si:H solar cell structure is shown in Fig. 1. The performance of

Table 1
Deposition condition for undoped μc-SiO_x:H thin films.

H_2/SiH_4	N_2O/SiH_4	Substrate temperature (°C)	Pressure (mTorr)	Power density (W)
20	0.2	200	250	110
26	0.2	200	250	110
30	0.2	200	250	110

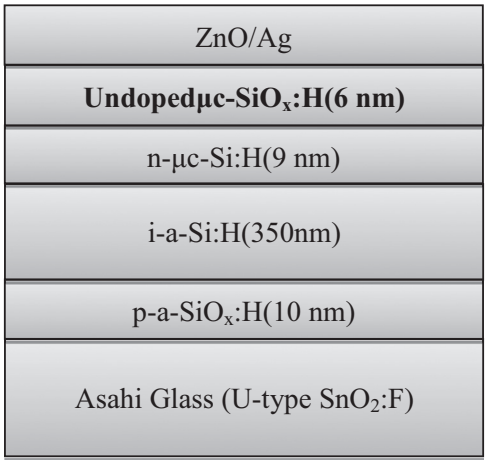

Fig. 1. Structure of superstrate-configuration p–i–n a-Si:H solar cells with an undoped μc-SiO_x:H layer.

the a-Si:H superstrate single-junction p–i–n thin-film solar cells was then defined according to J–V measurements obtained under standard AM1.5 (100 mW/cm^2) illumination at 25 °C. The external quantum efficiency (EQE) of the solar cells was measured at zero bias voltage to determine their spectral responses at various wavelengths. The J–V-tested cells had an active area of 1.0 cm^2.

3. Results and discussion

Fig. 2 shows the Raman scattering spectra of undoped μc-SiO_x:H films deposited at H_2/SiH_4 ratios of 20, 26, and 30, respectively. It can be seen clearly that these films revealed the presence of a mixture of μc-Si:H and amorphous silicon oxide phases[10]. The typical peak positioned at 521 cm^{-1} in bulk c-Si, reflecting a crystalline Si matrix[13]. When H_2/SiH_4 ratios gradually increases from 20 to 26, the Raman peak positions presents red shift from 512 to 518 cm^{-1}. However, when H_2/SiH_4 ratios at 30, the amorphous SiO_x:H film was found. This observation implies that the better crystallization of the Si-rich component in undoped μc-SiO_x:H films at H_2/SiH_4 ratios of 20 and 26 conditions and may provide better quality microcrystalline film compared to undoped amorphous SiO_x:H film prepared at H_2/SiH_4 ratios of 30.Fig. 3 shows the crystallinity and conductivity of the undoped μc-SiO_x:H films as the H_2/SiH_4 ratio was varied. Film crystallinity was evaluated using Raman scattering spectroscopy. We used the integrated area ratio

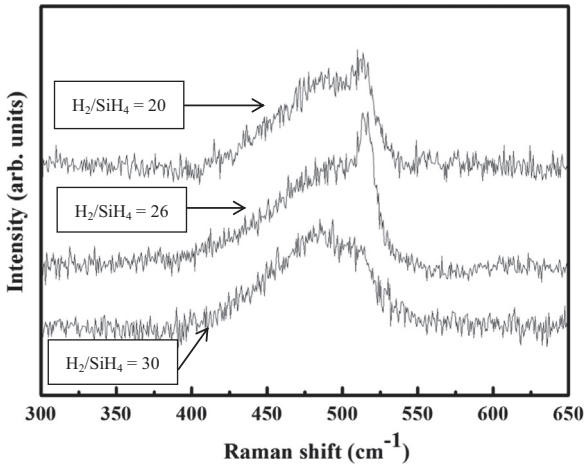

Fig. 2. Raman spectra of undoped μc-SiO_x:H thin films under different H_2/SiH_4 ratio.