Improved amorphous/crystalline silicon interface passivation for silicon heterojunction solar cells by hot-wire atomic hydrogen during doped a-Si:H deposition

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Abstract

Intrinsic/doped stacked hydrogenated amorphous silicon (a-Si:H) are widely used passivation layers for amorphous/crystalline silicon (a-Si/c-Si) heterojunction solar cells. This work reports that hot wire chemical vapor deposition of doped a-Si:H can significantly modify the property of the underlying intrinsic a-Si:H (a-Si:H(i)) as well as a-Si/c-Si interface passivation, which stems from the in-diffusion of highly reactive atomic hydrogen. Fourier transform infrared spectroscopy, spectroscopic ellipsometry and Raman analyses indicate that the underlying a-Si:H(i) films become more compact and less defected as a result of network reconstruction during doped a-Si:H capping. After this reconstruction, underdense a-Si:H(i) films obtained superior passivation quality than widely used dense layers, despite the inferior quality in the initial state. Effective minority carrier lifetime of c-Si passivated by underdense a-Si:H(i) was 19.9 ms, much higher than 15.2 ms in the case of using dense a-Si:H(i). The porous structure of underdense a-Si:H(i) facilitates hydrogen diffusion towards a-Si/c-Si interface and hence a rapid reduction of interface defect densities occurs, accounting for the better passivation quality. SHJ solar cells (160 μm , 156 $\times$ 156 mm²) with industry-compatible process were fabricated, yielding the efficiency up to 23.0% with high V_{oc} values of 741 mV.

Keywords

Surface passivation; Hydrogenated amorphous silicon; Amorphous/crystalline silicon heterojunction solar cells; Hot wire chemical vapor deposition

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1. Introduction

Among high-efficiency photovoltaic devices, amorphous/crystalline silicon heterojunction (SHJ) solar cells stand out due to their high performance and potential for low production cost [1]. A record conversion efficiency of 24.7% with an open circuit voltage (V_{oc}) of 750 mV was reported [2]. The doping of the hydrogenated amorphous silicon (a-Si:H) to form efficient p-n junction and back surface field with a high built-in voltage leads to high defect density in the a-Si:H bulk and at the a-Si/c-Si interface [3]. Therefore, a thin intrinsic passivation layer is inserted between the doped layers and wafer to reduce the defect density at the a-Si/c-Si interface (D_{it}) and thus restrain the surface carrier recombination, improving the V_{oc} of SHJ solar cells [4]. The typical 5–10 nm thick a-Si:H layers are usually prepared by chemical vapor deposition (CVD), either by plasma-enhanced CVD (PECVD) or by hot-wire CVD (HWCVD).

Post hydrogen plasma treatments in PECVD have been extensively applied in band gap engineering [5, 6], facilitating network crystallization [7], promoting amorphous-to-microcrystalline transition and improving the hydrogenation of wafer/film interface [8], via hydrogen in-diffusion from the kinetic plasma environment and further relaxation of the a-Si:H films. In contrast to the dissociation of H_2 by collisions with energetic electrons in PECVD, H_2 is efficiently decomposed into atomic H by catalytic cracking reaction with the heated filament in HWCVD [9]. Coincidentally, a calculated diffusion coefficient for hot-wire atomic hydrogen agrees well with the value for diffusion from hydrogen plasma [10], which implies the highly-reactive nature of hot-wire atomic H. These H atoms can penetrate into the as-deposited poly-Si_{1-x}Ge_x films and terminate the unsaturated dangling bonds,

with an effective post-hydrogenation depth up to ~250 nm [11]. Regarding this, the ultrathin (5-10 nm) intrinsic a-Si:H film for SHJ device must be susceptible to the exposure to hot-wire atomic hydrogen. As for doped a-Si:H deposition for SHJ device, a high hydrogen dilution (R_H) is commonly employed to enhance the crystallinity which benefits the doping efficiency [12]. This high R_H indeed leads to plentiful atomic H in the chamber, which may influence the underlying intrinsic films, a-Si/c-Si interface property and hence the passivation quality. The effect of inherent atomic H during the doped layer process in HWCVD has to be studied.

In this paper, we investigated the passivation effect induced by hydrogen penetration during subsequent deposition of n-type a-Si:H (a-Si:H(n^+)) in HWCVD. After a-Si:H(n^+) capping, underdense a-Si:H(i) film was observed to provide better passivation quality compared with widely used dense a-Si:H(i), despite the inferior passivation in the initial state. To clarify the improvement mechanism, a nanometer-accurate chemical etching method was carried out to uncover the microstructure evolution of underlying intrinsic films caused by the in-diffusion of highly-reactive atomic H during a-Si:H(n^+) capping. Finally, SHJ solar cells were optimized and a conversion efficiency of 23.0% was achieved. Such underdense a-Si:H(i) has great potential as an excellent passivation layer alternative for high performance SHJ solar cells.

2. Materials and methods

A-Si:H films were deposited on Czochralski phosphorus-doped c-Si (100) wafers (thickness, 160 μm ; resistivity, 1-6 $\Omega\text{ cm}$) in a multi-chamber HWCVD. The filaments for a-Si:H deposition are alloys which is composed of tungsten and tantalum [13]. Prior to a-Si deposition, the wafers were textured with potassium hydroxide, followed by the standard RCA cleaning process, then dipped in 2% diluted hydrofluoric acid (HF) for 1 min to remove the native oxide. A-Si:H(i) layers were deposited with pressure of 1.0 Pa, filament temperature (T_{fila}) of 1700 $^{\circ}\text{C}$, silane flow rates of 60 sccm without hydrogen dilution. For underdense a-Si:H(i), the substrate temperature (T_s) was set at 120 $^{\circ}\text{C}$, while dense a-Si:H(i) were fabricated at T_s of 160 $^{\circ}\text{C}$ (hereafter referred as I_{120} and I_{160} respectively). A-Si:H(n^+) film was deposited with T_s of 130 $^{\circ}\text{C}$, T_{fila} of 2000 $^{\circ}\text{C}$ and R_H of 4. In symmetrical i/ n^+ structure passivation as shown in Fig. 2, the thickness of a-Si:H(i) was fixed at 10 nm and that of a-Si:H(n^+) (d_{n^+}) varied, 4, 6, 8, 10 nm by adjusting deposition time. The lifetime of minority carrier (τ_{eff}) was measured

with micro-wave photo-conductance decay (μ -PCD) method (Sinton Consulting WCT-120 system, operated at the transient analysis mode and evaluated at a minority carrier injection density of $1.0 \times 10^{15} \text{ cm}^{-3}$).

To probe the structural evolution of the underlying intrinsic films after a-Si:H(n^+) deposition, we used a simple method of chemical etching, as illustrated in Fig. 1. 10 nm i, 6 nm n^+ /10 nm i and 10 nm n^+ /10 nm i for both I_{120} and I_{160} were deposited on single polished c-Si wafers (float-zone; thickness, 525 μm ; resistivity, 3000 $\Omega \text{ cm}$). The c-Si wafers were dipped in 2% HF to remove native oxide prior to a-Si:H deposition. After then, a-Si:H(n^+) overlayers were etched away in a mixed solution of 30% hydrogen peroxide (H_2O_2): 50% HF (4:1) with an average etching rate of $\sim 0.5 \text{ nm/min}$. More specifically, uppermost amorphous silicon is oxidized by H_2O_2 and the pre-formed oxide film is immediately etched away by HF. With spectroscopic ellipsometry, we verified the accurate etching to the i/n^+ interface by controlling etching time and no a-Si:H(n^+) traces remained on the sample (the remained a-Si:H thickness after etching is 10 nm, exactly that of a-Si:H(i) film). The optical parameters of a-Si:H films were characterized by Spectroscopic Ellipsometry (SE; J. A. Woollam, M-2000XI) fitted with Tauc-Lorentz model [14]. The Si-H_x bonding was characterized by Fourier Transform Infrared Spectroscopy (FTIR; Perkin Elmer, Spectrum100). For the FTIR characterization, a piece of the single polished Si wafer was dipped in 2% HF for 1 min to remove the surface native oxide, followed by a scan in the background mode. A-Si:H films were then characterized with FTIR in the sample mode. The complete transmittance spectra in the region $\omega = 450\text{-}4000 \text{ cm}^{-1}$ (see Fig. S1) were collected until CO_2 peak at 2350 cm^{-1} disappeared after N_2 blowing for 40 minutes to drain the air in the equipment. After then, the H bonding configuration was quantified by analyzing the stretching modes of Si-H_x bonding [15]. Raman spectra were recorded using a 325 nm He-Cd laser excitation (Jovin Yvon Labram 800002). For Raman characterization, thickness of a-Si:H(i) films were 40 nm by adjusting deposition time to absorb the laser completely so that the signal from c-Si substrate can be avoided. Cross-section images for a-Si/c-Si interface were acquired by high-resolution transmission electron microscopy (HR-TEM; Tecnai G2 F20 S-Twin), at a voltage of 200 kV.

FIG. 1. Schematic illustration of the characterization process of the underlying intrinsic a-Si:H by a simple chemical etching method.

3. Results and discussion

3.1 Passivation quality in i structure

Table 1 shows H incorporation, the maximum imaginary part ($\mathcal{E}_{i\text{-max}}$) of the dielectric function of as-deposited underdense/dense intrinsic films and the passivation quality in symmetrical a-Si:H(i)/c-Si(n)/a-Si:H(i) structure (i structure). Generally, low stretching mode (LSM, at 1980-2020 cm^{-1}) is assigned to isolated hydrogen in dense network structure, while high stretching mode (HSM, at 2070-2100 cm^{-1}) mainly represents clustered hydrogen at the internal surfaces of microvoids, corresponding to the poor film quality [15]. The microstructure factor R was calculated from the following equation [16]:

$$R = C_{\text{HSM}} / (C_{\text{HSM}} + C_{\text{LSM}})$$

Obviously, higher value of R represents higher porosity of a-Si:H structure. As Table 1 shows, HSM hydrogen content (C_{HSM}) predominates in hydrogen distribution for I₁₂₀ and correspondingly a high value of R was obtained, indicating a high degree of porosity associated with large amounts of strained Si-Si bonds and dangling bonds. This is due to a limited diffusivity of film-growth precursors and inadequate network reconstruction at low substrate temperature [17]. Raising T_s to 160 °C significantly reduces C_{HSM} while enlarges LSM hydrogen content (C_{LSM}), resulting in a significant reduction of R from 0.65 to 0.25. Meanwhile, hydrogen elimination occurs more readily due to adequate thermal hydrogen desorption [18], which accounts for the lower total hydrogen content (C_{total}) for I₁₆₀. In

addition, a pronounced increase in $\mathcal{E}_{i-\max}$ from 21.1 to 24.7 also demonstrates a larger mass density of I_{160} [19]. To summarize, I_{160} is more compact and less defective than I_{120} . Consequently, underdense a-Si:H(i) provided inferior passivation as evidenced by a very low τ_{eff} of 164 μs , which is almost one-eighth that of dense a-Si:H(i).

TABLE 1. Film properties and the passivation quality of 10 nm intrinsic layers in the as-deposited state.

	$C_{\text{HSM}} (\%)$	$C_{\text{LSM}} (\%)$	$C_{\text{total}} (\%)$	R	$\mathcal{E}_{i-\max}$	$\tau_{\text{eff}} (\mu\text{s})$
I_{120}	9.0	4.8	13.8	0.65	21.1	164
I_{160}	3.2	9.5	12.7	0.25	24.7	1327

3.2 Passivation quality in i/n⁺ structure

Contrary to the inferior performance in i structure passivation, underdense a-Si:H(i) performs better after a-Si:H(n⁺) capping. Figure 2 plots the τ_{eff} of c-Si symmetrically passivated by i/n⁺ stack layer versus incremental a-Si:H(n⁺) thickness. By increasing the thickness of a-Si:H(n⁺) layer to 10 nm, the τ_{eff} of c-Si passivated by I_{120} increases significantly from 0.164 ms to 19.9 ms, which is much higher than 15.2 ms in the case of using dense I_{160} . As Fig. 2 shows, in either case of intrinsic layers, τ_{eff} benefits substantially from thickening doped a-Si:H layers. Theoretically, a-Si:H(n⁺) layer induces a downward band bending inside the wafer, repelling the holes and hence reducing the electron/hole recombination at the a-Si/c-Si interface (field-effect passivation) [20]. Such thickness dependence of τ_{eff} is mainly due to the improved surface field effects which markedly gains with doped layer thickness. Note that since the field-effect passivation provided by doped layer is equal at each a-Si:H(n⁺) thickness in Fig. 2, the τ_{eff} difference between I_{120} and I_{160} must therefore derive from different D_{it} values. From the comparison, two distinct regions are identified. Initially ($d_{\text{n}^+} < 6$ nm), the higher τ_{eff} of I_{160} clearly indicates a lower D_{it} for dense passivation layer, consistent with τ_{eff} result in i passivation. Nevertheless, from about $d_{\text{n}^+} = 6$ nm onwards I_{120} obtains higher τ_{eff} , indicating a lower D_{it} for underdense intrinsic films after a-Si:H(n⁺) deposition despite the defective a-Si/c-Si interface in the initial state. This discrepancy indicates that the a-Si(i)/c-Si interface defect density is reduced during a-Si:H(n⁺) deposition. As the physical mechanism responsible for D_{it} reduction is the chemical passivation of Si- dangling bonds by hydrogen [21], it's inferred that subsequent a-Si:H(n⁺) deposition

induced rehydrogenation at the a-Si(i)/c-Si interface. This is the evidence of atomic hydrogen diffusion into the intrinsic films from the H-rich environment during a-Si:H(n^+) capping. As a consequence of longer periods of exposure to the atomic hydrogen by fabricating thicker a-Si:H(n^+), a higher hydrogenation level at the interface is obtained for underdense I_{120} compared with dense I_{160} , leading to better passivation performance. This demonstrates that the porous structure of underdense a-Si:H(i) facilitates hydrogen diffusion towards a-Si/c-Si interface and hence a more rapid reduction of D_{it} occurs.

FIG. 2. τ_{eff} (ms) values of c-Si passivated by i/ n^+ stack layer versus a-Si:H(n^+) thickness (d_{n^+}). Thickness of i was 10 nm and d_{n^+} varied, 4, 6, 8, 10 nm. For reference, values for i structure (depicted in Table I) is given as well (corresponding to $d_{n^+}=0$ nm).

3.3 Microstructure reconstruction of a-Si:H(i) during a-Si:H(n^+) capping

As Fig. 3 shows, subsequent n^+ deposition strongly affects the silicon-hydrogen bonding of the intrinsic films beneath. For both intrinsic films, the intensity of HSM markedly drops with increasing d_{n^+} while LSM absorbance increases, compared with the as-deposited state. On the one hand, large amounts of kinetic hydrogen atoms generated by dissociation of H_2 easily diffuse into a-Si:H(i) bulk and terminate the dangling bonds in the vacancies, which leads to the augment in C_{LSM} . On the other hand, as the H atoms move through the a-Si:H matrix, they either break or perturb strained Si-Si bonds [7]. Subsequent relaxation of these strained bonds results in the transformation of the film's structure and decreases the number of microvoids, as indicated by the decline of HSM absorbance. Consequently, the microstructure factor R decreases monotonically, i.e. from 0.25 to 0.06 for I_{160} as illustrated in Fig.

3(a)-(c) and from 0.65 to 0.44 for I_{120} in Fig. 3(d)-(f) (the error in determining R is shown in Fig. S2). Additionally, the imaginary part of dielectric function (ϵ_i) of a-Si:H(i) also increases as the arrow indicates in Fig. 4(a). This indicates the increscent mass density of a-Si:H(i) caused by H incorporation. Fig. 4(b) shows the Raman spectra of a-Si:H(i) films in the as-deposited state and after 10 nm n^+ depo&etch. The Raman signal due to the transverse optical mode was deconvoluted into three components: I_a (approx. 480 cm^{-1} , amorphous phase), I_m (approx. 510 cm^{-1} , crystalline phase with a grain size smaller than 10 nm) and I_c (approx. 520 cm^{-1} , crystalline phase with a grain size larger than 10 nm) [22]. On the one hand, as shown in Fig. 4(b), the single peak centered around 480 cm^{-1} suggests that the a-Si:H(i) films are purely amorphous without crystallization. On the other hand, the slight increase of the peak frequency after 10 nm a-Si:H(n^+) depo&etch for both I_{120} and I_{160} indicates a decrease in the network disorder [23] as well as a reduced lattice expansion [24]. This demonstrates that the underlying a-Si:H(i) becomes more ordered and compact as a result of network reconstruction caused by a mass of kinetic hydrogen during a-Si:H(n^+) capping.

FIG. 3. FTIR spectra evolutions of the 10-nm-thick intrinsic layers, deconvoluted into the LSM and HSM peaks with calculated microstructure factor R shown. (a) as-deposited (b) 6 nm n^+ deposition and etching (depo&etch) (c) 10 nm n^+ depo&etch for I_{160} and (d) as-deposited (e) 6 nm n^+ depo&etch (f) 10 nm n^+ depo&etch for I_{120} .

FIG. 4. (a) The imaginary part of dielectric function (ϵ_i) evolution and (b) Raman spectra evolution of a-Si:H(i) during a-Si:H(n^+) deposition.

Fig. 5(a) and Fig. 5(c) exhibits the cross-sectional HR-TEM images of a-Si:H(i)/c-Si for I_{120} and I_{160} . Pure silane deposition results in a typically amorphous bulk of a-Si:H(i), which is consistent with the single Raman peak centered around 480 cm^{-1} in Fig. 4(b). Meanwhile, a smooth a-Si(i)/c-Si interface without epitaxy was obtained, which is a key factor to yield outstanding surface passivation [25]. After 10 nm a-Si:H(n^+) deposition, the a-Si(i)/c-Si interface remains smooth for both I_{120} and I_{160} as displayed in Fig. 5(b) and Fig. 5(d). Additionally, no crystallite was observed in the highly-disordered a-Si:H(i) network, corresponding to the lack of I_c and I_m Raman peak after 10 nm a-Si:H(n^+) depo&etch in Fig. 4(b). To conclude, FTIR, SE, Raman and HR-TEM results demonstrate that a-Si:H(i) bulk property was enhanced and a smooth a-Si/c-Si interface was maintained during the kinetic hydrogen penetration during a-Si:H(n^+) capping. The enhanced a-Si:H(i) property reduces the carrier recombination in the bulk, partially contributing to the τ_{eff} improvement in Fig. 2.

It is noteworthy that the bulk property of I_{120} remains inferior than I_{160} even after reconstruction during 10 nm a-Si:H(n^+) deposition as revealed by the greater R (0.44 versus 0.06) displayed in Fig. 3(f) and Fig. 3(c) and the smaller ϵ_i in Fig. 4(a), which seems contradictory to the higher τ_{eff} (19.9 ms vs. 15.2 ms) as shown in Fig. 2. The poorer bulk property yet higher τ_{eff} for underdense films demonstrates that a-Si/c-Si interface quality rather than a-Si:H bulk property determines c-Si surface passivation quality. Such underdense a-Si:H(i) capped by a-Si:H(n^+) has the potential to enhance the V_{oc} of SHJ solar cell thanks to the higher a-Si/c-Si interface hydrogenation level.

FIG. 5. HRTEM image of c-Si coated by (a) 10 nm I_{120} , (b) 10 nm $I_{120}/10$ nm n^+ , (c) 10 nm I_{160} and (d) 10 nm $I_{160}/10$ nm n^+ .

3.4 Cell performance

Based on the discussion above, 160 μm SHJ rear emitter solar cells ($156 \times 156 \text{ mm}^2$) were fabricated with industry-compatible process, details about the cell structures and process can be found in our previous paper [26]. The dependence of current-voltage parameters of SHJ solar cells versus a-Si:H(n^+) thickness is illustrated in Fig. 6, measured at standard condition of AM1.5 and 25 °C. As Fig. 6(a) shows, V_{oc} sustainably increases with d_{n^+} , consistent with τ_{eff} results as shown in Fig. 2. The higher V_{oc} of cell passivated by I_{120} compared with I_{160} further confirms the lower D_{it} and better passivation due to more adequate rehydrogenation at the a-Si/c-Si interface [27], as a result of the in-diffusion of hot-wire atomic hydrogen. From the viewpoint of c-Si surface passivation, thicker a-Si:H(n^+) layer is a positive factor for SHJ solar cell performance but this in turn leads to more parasitic absorption [28] and thus lower J_{sc} as shown in Fig. 6(b). Nevertheless, it is interesting to note the improvement in J_{sc} using underdense a-Si:H(i) as front-side passivation layers thanks to the lower parasitic optical loss [29], as implied by lower $\mathcal{E}_{i-max}$ and higher C_H in Table 1. Therefore, such underdense intrinsic passivation layer provides an approach towards higher J_{sc} of SHJ cells without deteriorating electronic passivation quality by HWCVD. Although the loose network of I_{120} results in higher series resistance as revealed by the lower FF in Fig. 6(c), the loss in FF can be offset by the

gain in V_{oc} and J_{sc} . At an optimal a-Si:H(n^+) thickness of 8 nm, a high conversion efficiency up to 23.0% with V_{oc} of 741 mV was achieved.

FIG. 6. (a) V_{oc} , (b) J_{sc} , (c) FF, and (d) Eff. of SHJ solar cells versus a-Si:H(n^+) thickness, squares for dense I₁₆₀ and circles for underdense I₁₂₀.

4. Conclusions

To summarize, hot-wire atomic hydrogen during subsequent n-type a-Si:H deposition can significantly modify the microstructure and improve the bulk property of the underlying intrinsic passivation layers, experimentally investigated by a nanometer-accurate chemical etching method. Additionally, c-Si surface passivation quality was improved as a result of kinetic hydrogen in-diffusion towards the a-Si/c-Si interface. Compared with widely used dense a-Si:H(i), a higher hydrogenation level at the a-Si/c-Si interface was obtained for underdense a-Si:H(i) as the porous structure facilitates hydrogen diffusion. Consequently, higher τ_{eff} and higher V_{oc} of SHJ solar cells was obtained, demonstrating that such underdense a-Si:H(i) capped by doped a-Si:H has great potential as an excellent passivation layer alternative for high performance SHJ solar cells. Our results revealed that doping layer deposition in HWCVD not only introduces field effect but also may improve the chemical passivation quality of the underlying intrinsic films. This concept is promising for guiding the design

of intrinsic/doped a-Si:H stack layers in pursuit of high efficiency of SHJ solar cells.

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