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Hydrogen concentration at a-Si:H/c-Si heterointerfaces—The impact of deposition temperature on passivation performance

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ABSTRACT
We studied the effect of deposition temperature on the hydrogen distribution and the passivation performance of hydrogenated amorphous silicon (a-Si:H) coated crystalline silicon (c-Si) heterojunctions as a model of high efficiency solar cell structures. Nuclear reaction analysis (NRA) was employed to obtain hydrogen depth profiles of the heterojunctions prepared at temperatures from 80 to 180 °C. The implied open circuit voltage (i-VOC) and carrier lifetime monotonically increased with increasing deposition temperature in the as-deposited samples. NRA clarified that the hydrogen concentration (CH) at the a-Si:H/c-Si interface and in the a-Si:H layer decreased with deposition temperature. The hydrogen concentration around the interface was roughly 3×10^{21} cm^{-3} for the sample deposited at 180 °C. The NRA results are supplemented by optical constants obtained with spectroscopic ellipsometry (SE). At higher growth temperature, larger refractive indices and extinction coefficients were confirmed by SE analysis, suggesting that fewer hydrogen atoms are incorporated into the a-Si:H layers prepared at higher growth temperature. Furthermore, the passivation performance was enhanced by post deposition annealing (PDA) at 200 °C for 30 min. No significant change of the hydrogen distribution and optical constants was observed after PDA, suggesting that improved passivation is due to a local rearrangement of hydrogen at the molecular level that results in enhanced hydrogenation of dangling bonds.

I. INTRODUCTION
Silicon heterojunction (SHJ) solar cells employing hydrogenated amorphous silicon (a-Si:H) have garnered significant attention owing to their extremely high conversion efficiency.1–4 The high conversion efficiency is primarily due to the passivation of the crystalline silicon (c-Si) surface by intrinsic type a-Si:H (i-a-Si:H), which suppresses carrier recombination at the c-Si surface and thus allows for a high open circuit voltage (VOC).1–3 Korte et al. proposed that two recombination pathways of photogenerated carriers exist in a-Si:H passivated c-Si: recombination of carriers via dangling bonds at the a-Si:H/c-Si heterointerface and in the bulk of a-Si:H.3 According to the description of the surface recombination mechanism, the interface recombination rate depends on the interface state density, which is proportional to the density of interfacial dangling bonds.4 A low density of interfacial dangling bonds is believed to arise from hydrogenation of Si dangling bonds.4–13 The hydrogenation of dangling bonds around a-Si:H/c-Si heterointerfaces is regarded as the origin of the passivation effect, however there are few reports on the correlation between passivation performance and hydrogen distribution around a-Si:H/c-Si heterointerfaces. Thus, the quantitative investigation of the hydrogen distribution near the
a-Si:H/c-Si interface is necessary for a fundamental comprehension of the passivation mechanism by a-Si:H and for further device optimization.

High quality a-Si:H/c-Si heterointerfaces and a-Si:H layers are essential to achieve high efficiency SHJ solar cells. 7,14 Generally, the quality of a-Si:H films strongly depends on the deposition conditions, particularly on the deposition temperature. Higher deposition temperatures reduce the number of dangling bonds at the a-Si:H/c-Si interface 7 and in a-Si:H, 7 resulting in improvement of the passivation performance. Meanwhile, crystallization of a-Si:H occurs above a critical deposition temperature, deteriorating the passivation performance. 7,16,17 Knowing the hydrogen concentration near the a-Si:H/c-Si interface is critical to clarify the mechanism of the hydrogen passivation and its influence on the solar cells properties. So far, the hydrogen distribution has been studied using Secondary Ion Mass Spectroscopy (SIMS). 7 In SIMS measurements, however, such as the knock-on effect cannot be neglected and hence the depth profiles become obscure. 18,19 Furthermore, hydrogen bonding state in depth was investigated by a combination of Spectroscopic Ellipsometry (SE) and Fourier Transform Infrared Spectroscopy (FTIR) in Attenuated Total Reflectance (ATR). 20,23 Although a lot of efforts is contributed to clarify the passivation effect, the impact of deposition temperature on the passivation performance of a-Si:H/c-Si and its interfacial hydrogen distribution has not yet been presented. Hence, the correlation between the passivation performance and the hydrogen concentration near the interface has remained not yet fully understood.

In this paper, we studied the influence of the deposition temperature on the passivation performance and the hydrogen depth distribution in a-Si:H/c-Si heterojunctions with SE and hydrogen depth profiling by Nuclear Reaction Analysis (NRA). The passivation performance of the a-Si:H/c-Si heterojunctions is found to be improved with increasing deposition temperature. The NRA measurements clarify that lower concentrations of hydrogen in the bulk of the a-Si:H films and at the a-Si:H/c-Si interface are realized in highly passivated a-Si:H/c-Si heterojunctions. The SE analysis reveals that the layer thickness of the a-Si:H films decreases with increasing deposition temperature, while the refractive index (n) and the extinction coefficient (k) increase. Moreover, almost identical hydrogen distributions and n-k spectra are observed for as-deposited and annealed a-Si:H/c-Si heterojunctions.

II. EXPERIMENTAL METHODS

All samples were prepared by a Plasma Enhanced Chemical Vapour Deposition (PECVD) with a frequency of 27.12 MHz (ULVAC Inc., CME-200J). Double side mirror-polished, float zone (FZ) grown, 280-μm-thick n-type c-Si(100) (n-c-Si) substrates were used. Prior to deposition of a-Si:H, the substrates were cleaned by Semicoolean-23 (Furuuchi Chemicals) for 6 minutes in an ultrasonic bath. The native oxide on the substrates was removed by immersing the substrates into 2.5% HF for 1 minute and then dipping into ozoneing, deionized water (DI-O3) for 10 minutes to clean their surfaces and form a protective oxide layer. 20–22 The concentration of O3 in DI-O3 was about 25 ppm. Subsequently, the protective oxide layer was stripped off by 2.5% HF for 1 min, and the substrates were quickly loaded into the PECVD chamber. The i-a-Si:H layers were deposited on c-Si substrates with a silane (SiH4) flow rate of 40 sccm, total pressure of 25 Pa and RF power density of 32.5 mW/cm2. The deposition temperature (Tdep) varied from 80 to 205 °C. The deposition time of i-a-Si:H tdep was fixed at 115 s. Furthermore, Post-Deposition Annealing (PDA) was carried out in air using a hot plate at 200 °C for 30 min. The PDA condition used in typical screen printing metallization for SHJ solar cells is employed. 23,24

The layer thickness of a-Si:H (tdep) was measured by SE (J. A. Woollam Co., M-2000DI-Nug). The Tauc-Lorentz model was used as an optical model. A native oxide layer with a thickness of about 1 nm was assumed in the SE analysis. To model the native oxide layer, the optical constant of thermally grown silicon oxide was used. The optical constant was determined by analyzing the 25-nm-thick thermally grown silicon oxide. 25 The layer thickness of the oxide was not fixed in the analysis, while the optical constant was unchanged. The values of mean square error for all samples ranged from 1.2 to 1.5, indicating that the optical model is reasonable. For characterization of the passivation performance, effective carrier lifetime (τeff) and implied VOC (i-VOC) was measured by quasi-steady-state photoconductance (Sinton Instruments, WCT-120) in generalized 1/1 and 1/64 modes. 25–27 Hydrogen depth profiles were obtained by NRA via 1H(12N,α)12C. The NRA technique utilizes the nuclear reaction between high energy 15N ions and hydrogen in the sample to non-destructively quantify the hydrogen concentration at buried interfaces. 33,34 The resonant 1H(12N,α)12C nuclear reaction has a narrow energy resonance at 6.385 MeV and emits γ-rays with a specific energy of 4.43 MeV. The γ-ray yield is proportional to the number of incident 15N ions and to the hydrogen concentration (C12) in the sample. The incident high energy 15N ions gradually lose kinetic energy while penetrating into the sample and react with hydrogens at the depth where the resonance energy is met. Therefore, hydrogen depth profiling is achieved by collecting the γ-ray yield while changing the energy of incident 15N projectile ions. Further details of the NRA technique are given elsewhere. 33–35 The 15N2+ ion beams of about 1 × 1013 A were generated by the MALT van de Graaff tandem accelerator at the University of Tokyo. 35 The γ-rays were collected by a scintillation detector. To quantify the H content, the γ-detection sensitivity was calibrated with Kapton film as H concentration standard. 33,35 The Kapton standard has a density of 1.45 g/cm3, a H-concentration of 2.28 × 1022 cm−3 and a Stopping Power (STP) of 1.2879 keV/nm. The calibration measurement resulted in a sensitivity constant of α = 1.89 × 1019 H/cm3. H concentrations are calculated from the NRA γ-yields (Y) as C12 = Y × STP/α. The error bar for α (in the order of 5% or less) and the statistics for Y will determine the error bar for the H concentration. 35 The STP values of 1.4406, 1.4303, 1.4265, 1.4254, and 1.4244 keV/nm were used for the samples deposited at 80, 110, 140, 160, and 180 °C, respectively, reflecting their different H content. The STP values are calculated based on Bragg’s rules. 35 Firstly, the H concentration from the Y was roughly estimated assuming the STP of pure Si, then Bragg’s rule was used to derive a more appropriate STP value by considering in addition the maximum H content in the film. The position of the a-Si:H/c-Si interface was determined by SE analysis. Furthermore, hydrogen bonding states were investigated by the FTIR employing ATR method. The ATR-FTIR measurements was carried out by using a ZnSe prism in N2 atmosphere.
III. RESULTS AND DISCUSSION

Figure 1 shows $\tau_{\text{eff}}$ curves of (a) as-deposited and (b) annealed i-a-Si:H/n-c-Si heterojunctions prepared at $T_{\text{depo}}$ from 80 to 205 °C as a function of minority carrier density. Figure 2 shows the influence of $T_{\text{depo}}$ on (a) $t_{\text{a-Si:H}}$, (b) $\tau_{\text{eff}}$ and (c) $i-V_{\text{OC}}$ of as-deposited and annealed i-a-Si:H/n-c-Si heterojunctions. The values of $t_{\text{a-Si:H}}$ and $i-V_{\text{OC}}$ before and after PDA are summarized in Table I. Decreased $\tau_{\text{eff}}$ and $i-V_{\text{OC}}$ values were observed for the i-a-Si:H/n-c-Si heterojunctions fabricated at $T_{\text{depo}} = 205$ °C. It is reported that the deterioration of the passivation performance at high $T_{\text{depo}}$ is caused by crystallization of the a-Si:H layer and crystallization is promoted by PDA.\textsuperscript{11,16,17} From Table I, an increase in passivation performance is observed at $T_{\text{depo}} = 80\sim180$ °C after PDA, suggesting no crystallization occurred for the as-deposited samples. The $t_{\text{a-Si:H}}$ decreases with increasing with $T_{\text{depo}}$ from 80 to 180 °C, while the $\tau_{\text{eff}}$ and $i-V_{\text{OC}}$ increase. Note that the $t_{\text{a-Si:H}}$ is 115 s for all samples in Fig. 1 and 2 and only $T_{\text{depo}}$ is varied. The reduction of $t_{\text{a-Si:H}}$ is mainly caused by the reaction of precursors with the sample surface. In general, the deposition process of a-Si:H consists of the following processes: generation of precursors, transportation of precursors to the surface, and deposition by reaction of precursors on the surface.\textsuperscript{36} The surface migration of precursors is enhanced at higher $T_{\text{depo}}$ and thus a more compact i-a-Si:H film is formed. The improved passivation effect can be attributed to a reduced number of nano-voids in the bulk of i-a-Si:H and at the i-a-Si:H/n-c-Si interface. The existence of void structures in a-Si:H has been reported previously in studies by infrared spectroscopy,\textsuperscript{37} positron annihilation,\textsuperscript{38} nuclear magnetic resonance,\textsuperscript{39} Small-Angle X-ray Scattering (SAXS),\textsuperscript{40} and SE.\textsuperscript{41} Mahan \textit{et al.} confirmed that the density of voids in a-Si:H layer decreases with increasing substrate temperature by SAXS.\textsuperscript{40} The nano-voids around a-Si:H/c-Si interfaces increase the vacancies around the interface.\textsuperscript{9,42} Consequently, the unpassivated dangling bonds increased, resulting in increased carrier recombination.

Figure 3 shows NRA γ-ray yield curves measured for as-deposited i-a-Si:H/n-c-Si heterojunctions prepared at $T_{\text{depo}}$ of 80, 110, 140, 160 and 180 °C as a function of the incident $^{15}$N$^+$ ion energy. The vertical black and coloured solid lines in Fig. 3 indicate the surface and i-a-Si:H/n-c-Si interface positions of each sample, which were determined from the layer thicknesses obtained in the SE measurements. It is seen that inflection points very closely agree with the film thicknesses determined by SE. Using the STP values, interface positions can be directly calculated from the $^{15}$N energies at the respective interfacial inflection points. With increasing $T_{\text{depo}}$ from 80 to 180 °C, $C_{\text{H}}$ in the bulk of the a-Si:H layer decreases from about $8 \times 10^{21}$ to $5 \times 10^{21}$ cm$^{-3}$. During growth of a-Si:H, excess H atoms are released and reaches steady state values of the bulk a-Si:H after the precursors stick to the growing surface.\textsuperscript{43} Hence, we suppose that desorption of hydrogen from the growing a-Si:H surface is enhanced at higher $T_{\text{depo}}$ and thus more compact a-Si:H layers were produced. The values of $C_{\text{H}}$ at the interface before and after PDA are given in Table I. The $C_{\text{H}}$ at the i-a-Si:H/n-c-Si interface tends to decrease with increasing $T_{\text{depo}}$ from 80 to 180 °C. Furthermore, the passivation performance of the i-a-Si:H/n-c-Si heterojunctions was...
improved by employing higher $T_{\text{depo}}$. These results indicate that the excellent passivation performance correlates with fewer hydrogens at the interface. It should be noted again that deposition conditions are constant except $T_{\text{depo}}$. The generation of precursors and transportation of precursors to the surface would be almost same for all samples. The surface migration and the desorption of hydrogen can be dependent on $T_{\text{depo}}$ and be enhanced during deposition as $T_{\text{depo}}$ increases. Therefore, we deduced that excessive hydrogens are desorbed from the growing a-Si:H surface and are more efficiently consumed for hydrogenation of dangling bonds during growth at higher $T_{\text{depo}}$.

The hydrogen bonding states were investigated by ATR-FTIR measurements. Figure 4 (a) shows ATR-FTIR spectra of i-a-Si:H/n-c-Si grown at 80, 110, 140, 160 and 180 °C. FTIR peaks around 2000 cm$^{-1}$ were observed for all samples. In general, the peak of 2000 cm$^{-1}$ represent isolated hydride (SiH), while the peak of 2100 cm$^{-1}$ arises from clustered monohydride and polyhydride (SiH$_n$) stretching mode. The hydrogen bonding states were investigated by the ratio $R$ of the peak intensity at 2100 cm$^{-1}$ ($I_{2100}$) to total intensity at 2000 and 2100 cm$^{-1}$ ($I_{2000} + I_{2100}$), i.e. $R = I_{2100}/(I_{2000} + I_{2100})$. Figure 4 (b) shows the dependence of $R$ on $T_{\text{depo}}$. The dotted line in Fig. 4 (b) is a visual guide. The bonding states of SiH become dominant as $T_{\text{depo}}$ increased, indicating less hydrogens incorporated in the a-Si:H, which is good agreement with results of NRA. It is reported that isolated monohydrides incorporate into a-Si:H layers with lower $C_H$ whereas clustered monohydrides and dihydrides reside in a-Si:H layers with higher $C_H$. Therefore, reduced hydrogen incorporation and a more compact a-Si:H layer is expected to be obtained at higher $T_{\text{depo}}$, which is in good agreement with the NRA results. The densification of bulk a-Si:H layer with $T_{\text{depo}}$ is additionally supported by the SE analysis. Figure 5 shows (a) the refractive index ($n$) and (b) extinction coefficient ($k$) of the as-deposited i-a-Si:H/n-c-Si heterojunctions prepared at $T_{\text{depo}}$ of 80, 110, 140, 160,

| $T_{\text{depo}}$ [°C] | $t_{\text{a-Si:H}}$ [nm] | $i-V_{\text{OC}}$ [V] | $C_H$ at a-Si:H/c-Si interface [cm$^{-3}$] |
|------------------------|--------------------------|--------------------------|--------------------------------------------|
|                        | as-deposited | annealed | as-deposited | annealed | as-deposited | annealed |
| 80                     | 34.5        | 34.4     | 0.563        | 0.622    | 6.0 $\times$ 10$^{21}$ | 5.1 $\times$ 10$^{21}$ |
| 110                    | 22.3        | 22.4     | 0.653        | 0.718    | 3.6 $\times$ 10$^{21}$ | 3.6 $\times$ 10$^{21}$ |
| 140                    | 19.7        | 20.1     | 0.707        | 0.714    | 3.5 $\times$ 10$^{21}$ | 3.2 $\times$ 10$^{21}$ |
| 160                    | 19.7        | 19.4     | 0.707        | 0.709    | 3.4 $\times$ 10$^{21}$ | 2.9 $\times$ 10$^{21}$ |
| 180                    | 18.7        | 18.4     | 0.722        | 0.724    | 3.0 $\times$ 10$^{21}$ | 3.0 $\times$ 10$^{21}$ |
| 205                    | 15.7        | 15.5     | 0.709        | 0.680    | -                  | -                |

Figure 3. Hydrogen depth profiles of as-deposited i-a-Si:H/n-c-Si heterojunctions prepared at different $T_{\text{depo}}$ by NRA. The vertical black and coloured solid lines indicate the surface and i-a-Si:H/n-c-Si interface of each sample. The position of the i-a-Si:H/n-c-Si interface was determined by SE analysis.

Figure 4. Deposition temperature dependence of (a) FTIR absorbance spectra of the as-deposited a-Si:H on c-Si and (b) ratio of peak intensity at 2100 cm$^{-1}$ to total intensity at 2000 and 2100 cm$^{-1}$. The dotted line in Fig. 4 (b) is a visual guide.
180 and 205 °C as a function of wavelength. With increasing $T_{\text{depo}}$, the values of $n$ and $k$ increase. The increase in $n$ and $k$ is ascribed to fewer hydrogen species in the silicon matrix.\(^{41,45}\) Furthermore, the hump of $n$ and $k$ can be seen at around 400 nm at $T_{\text{depo}} = 205$ °C, which is arisen from the c-Si. Therefore, higher passivation performance of i-a-Si:H/n-c-Si correlates with less $C_H$ at the interface and more compact i-a-Si:H layers without crystallization of a-Si:H.

After PDA at 200 °C for 30 min, almost identical $\tau_{\text{eff}}$ compared to as-deposited samples was observed, whereas increased $\tau_{\text{eff}}$ and $i-V_{\text{OC}}$ were observed in Fig. 2 and Table I. These results suggest that PDA induced no significant structural change in bulk i-a-Si:H while recombination sites are reduced. Figure 6 shows the NRA H profiles of i-a-Si:H/n-c-Si heterojunctions at $T_{\text{depo}} = (a)$ 180, (b) 160, (c) 140, (d) 110 and (e) 80 °C before and after PDA. The differences of $C_H$ in Fig. 6 are small, however, a slight variation of $C_H$ can be observed. In particular, $C_H$ at the i-a-Si:H/n-c-Si interface at $T_{\text{depo}} = 80$ °C was noticeably reduced. Shultz et al. confirmed that no bulk a-Si:H reconfiguration takes place by PDA. Furthermore, De Wolf et al. remarked that a rearrangement of hydrogens around a-Si:H/c-Si heterointerfaces by PDA is responsible for the enhancement of passivation performance.\(^{11}\) It is considered that the rearrangement is caused by a transition from a non-equilibrium to an equilibrium structure.\(^{11}\) Accordingly, the enhancement of passivation performance by PDA can be attributed to hydrogenation of dangling bonds by a rearrangement of hydrogens around a-Si:H/c-Si interface.

Figure 7 shows $n$ and $k$ of annealed i-a-Si:H/n-c-Si heterojunctions prepared at $T_{\text{depo}}$ of 80, 110, 140, 160 and 180 °C. Almost identical spectra were obtained for both as-deposited and annealed i-a-Si:H/n-c-Si heterojunctions. At $T_{\text{depo}} = 80$ °C, the value of $n$ was increased after PDA, indicating densification of a-Si:H. Fujiwara et al. investigated growth kinetics of a-Si:H by in situ SE and ATR-FTIR and clarified that the SiH$_2$ bonding state is dominant at the early stage of a-Si:H growth.\(^{18}\) Hence, excessive hydrogens exist around the i-a-Si:H/n-c-Si interface and effusion of hydrogen is induced by PDA, which results in reduced $C_H$ around the interface and increase in $n$.

From these results we conclude that improved passivation performance is obtained in case of a moderate hydrogen concentration at the a-Si:H/c-Si interface. Denser a-Si:H layers are formed possibly due to the enhanced surface migration of precursors and desorption
of hydrogen from the growing a-Si:H surface at higher $T_{\text{depo}}$, which leads to efficient hydrogenation of dangling bonds and densification of the a-Si:H layer. Consequently, small quantities of hydrogen at the interface and high value of $n$-$k$ is observed for highly passivated i-a-Si:H/n-c-Si heterojunctions in comparison to poorly passivated the heterojunctions. Furthermore, the passivation performance was enhanced by PDA, even though no significant change of the hydrogen depth profile was observed. Therefore, the enhanced passivation performance appears to correlate with only a local rearrangement of hydrides leading to a more complete hydrogenation of dangling bonds around the i-a-Si:H/n-c-Si heterointerfaces. Finally, it is important to note that the plasma excitation frequency was fixed at 27.12 MHz in this work. The deposition rate and electrical properties are dependent on plasma excitation frequency, since dissociation of silane relies on plasma excitation frequency. Except the dissociation process, the deposition processes of a-Si:H using other plasma excitation frequency may be almost same as our experiments. Therefore, our results would partly valid for different plasma excitation frequency.

IV. CONCLUSIONS

We studied the impact of growth temperature on the relationship between the $C_H$ distribution in i-a-Si:H/c-Si heterojunctions and their passivation performance by NRA H depth profiling. The passivation performance reached a maximum at $T_{\text{depo}}$ of 180 °C for as-deposited and annealed samples. The $C_H$ depth profile indicates that reduced $C_H$ in the i-a-Si:H layer, that is a denser a-Si:H layer, is realized at higher $T_{\text{depo}}$. Possibly due to enhanced surface migration of precursor molecules and desorption of hydrogen. In addition, the $C_H$ of $\sim 3 \times 10^{17} \text{ cm}^{-3}$ at the i-a-Si:H/n-c-Si interface leads to highly passivated c-Si. While the $C_H$ depth profiles remained nearly identical after PDA, passivation performance was enhanced, suggesting that enhanced hydrogenation of dangling bonds is caused by local rearrangements of hydrogen species. We conclude that the requirement for highly passivated c-Si is a compact a-Si:H layer as well as a moderate hydrogen concentration in the order of $3 \times 10^{17} \text{ cm}^{-3}$ near the a-Si:H/c-Si interface.

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