Silicon Nanowire Heterojunction Solar Cells with an Al₂O₃ Passivation Film Fabricated by Atomic Layer Deposition

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Abstract
Silicon nanowires (SiNWs) show a great potential for energy applications because of the optical confinement effect, which enables the fabrication of highly efficient and thin crystalline silicon (c-Si) solar cells. Since a 10-μm-long SiNW array can absorb sufficient solar light less than 1200 nm, the 10-μm-long SiNW was fabricated on Si wafer to eliminate the influence of the Si wafer. On the other hand, Surface passivation of the SiNWs is a crucial problem that needs to be solved to reduce surface recombination and enable the application of SiNWs to c-Si solar cells. In this study, aluminum oxide (Al₂O₃) was fabricated by atomic layer deposition for the passivation of dangling bonds. However, owing to a complete covering of the SiNWs with Al₂O₃, the carriers could not move to the external circuit. Therefore, chemical–mechanical polishing was performed to uniformly remove the oxide from the top of the SiNWs. A heterojunction solar cell with an efficiency of 1.6% was successfully fabricated using amorphous silicon (a-Si). The internal quantum efficiencies (IQE) of the SiNW and c-Si solar cells were discussed. In the wavelength region below 340 nm, the IQE of the SiNW solar cell is higher than that of the c-Si device, which results in an increase of the absorption of the SiNW cells, suggesting that SiNWs are promising for crystalline-silicon thinning.

Keywords: Silicon nanowire, Passivation, Chemical–mechanical polishing, Atomic layer deposition, Solar cell

Introduction
Crystalline silicon (c-Si) solar cells are widely used worldwide because of their high efficiency and abundance [1–9]. To reduce the power generation costs of such solar cells, their efficiency must be increased and their fabrication cost must be reduced. However, the efficiency of c-Si solar cells is close to the theoretical efficiency limit and further improvement is difficult because the open-circuit voltage (V_{oc}) is limited by Auger recombination [10, 11]. Creating very thin c-Si films is an effective way to improve V_{oc}, but extremely thin c-Si solar cells exhibit a low short-circuit current density (I_{sc}) because of their low absorption coefficient [12, 13]. Recently, silicon nanowires (SiNWs) have attracted considerable attention because they exhibit a strong optical confinement effect that is essential for trapping light in solar cells [14–21]. In our previous experiments, we succeeded in evaluating the optical properties of SiNWs by peeling them from silicon wafers using polydimethylsiloxane [22]. A 10-μm-long SiNW array can absorb sufficient light, which indicates that SiNWs can reduce the thickness of c-Si solar cells. Since it is difficult to fabricate self-standing SiNW array, the Si wafer is needed. In this study, we focused on the fabrication of 10-μm-long SiNW arrays on the Si wafer. Therefore, to maximize the absorption in the wavelength below 1200-nm region by 10-μm-long SiNW arrays, the influence of Si wafer can be eliminated. On the other hand, to apply SiNWs to solar-cell structures, it is necessary to fabricate a passivation film on their surface to reduce surface recombination. We found that SiNWs exhibit a high aspect ratio, so it is difficult to fabricate a passivation film by chemical vapor deposition. Therefore, the passivation film was fabricated on the SiNW surface by atomic layer deposition (ALD) [23, 24]. On the other hand, SiNW arrays containing Al₂O₃ cannot be peeled
from the silicon wafer due to the improved mechanical strength. Moreover, the carriers cannot move to the external circuit because of the insulating Al$_2$O$_3$ film. In this study, we propose a new structure (shown in Fig. 1) in which 10-μm-long SiNWs are fabricated on a Si wafer.

To form a contact between the SiNWs and a-Si, the Al$_2$O$_3$ present on the top of the SiNWs was removed by chemical–mechanical polishing (CMP) and etching. The influence of Al$_2$O$_3$ etching on the properties of the solar cells was investigated.

**Methods**

**Fabrication of SiNW Arrays and Al$_2$O$_3$**

A p-type Si (100) wafer (8–10 Ω cm, 550 μm) was immersed in hydrofluoric acid (HF) solution with AgNO$_3$ to deposit silver particles. The Si wafer was chemically etched, using 4.8 M HF and 0.15 M H$_2$O$_2$ at room temperature, and subsequently added into an HNO$_3$ solution to remove the silver films. Finally, the oxide layer present on the prepared SiNW array was removed using the HF solution. SiNWs with lengths of 10, 15, and 20 μm were fabricated by changing the etching time. Since the space between the SiNWs is large, silica particles with a diameter of about 80 nm (dispersed in an ethanol solution) were filled into the space between the wires. Then, 66-nm-thick Al$_2$O$_3$ was deposited by ALD to passivate the dangling bonds. Field-emission scanning electron microscopy (FE-SEM, JEOL JSM-7001F) was applied to examine the structure of the prepared SiNW arrays.

**Removal of Al$_2$O$_3$ on the Top of SiNWs**

Next, an etching paste and the CMP method were applied to remove the top of the SiNWs and the Al$_2$O$_3$ on them. Figure 2a shows the Al$_2$O$_3$ etching procedure using an etching paste. The etching paste was formed on the Al$_2$O$_3$ layer, followed by annealing to remove it. Finally, the etching paste was removed. In the case of CMP, the detailed process is shown in Fig. 2b. With the fabricated solar cell structure, the length of the SiNW array remained constant at 10 μm, and therefore, the etching thickness was changed by changing the initial length of the SiNW arrays. When the initial length of the SiNWs was 10 μm, the etching was stopped at the top of the nanowires (etching thickness 0 μm, the length of SiNW 10 μm, the thickness of remaining Si wafer 540 μm), which means that the Al$_2$O$_3$ above the SiNWs was only etched. For an initial SiNW length of 15 μm, the etching length was defined as 5 μm, including the 5-μm SiNWs and Al$_2$O$_3$ (etching thickness 5 μm, the length of SiNW 10 μm, the thickness of remaining Si wafer 535 μm). When the etching length was defined as 10 μm, the initial length was 20 μm (etching thickness 10 μm, the length of SiNW 10 μm, the thickness of remaining Si wafer 530 μm).

**Fabrication of the Solar Cell Structure**

Figure 1 shows the solar-cell structures fabricated herein; the heterojunction structure of a-Si and Si was adopted. Fabrication procedure and condition of the heterojunction structure are the same as the SiNW solar cell structure with Al$_2$O$_3$.
In the case of reference solar cells, the p-type Si (100) wafer (8–10 Ω cm, 550 μm) was used without SiNW. A double heterojunction was formed by depositing an i-type hydrogenated amorphous silicon layer (i-a-Si:H, thickness 5 nm), an n-type a-Si:H layer (thickness 10 nm), and a p-type a-Si:H layer (thickness 10 nm) via plasma-enhanced chemical vapor deposition (PECVD). Indium tin oxide (ITO) (thickness 80 nm) and an Ag grid were used to fabricate the front electrode. The reflectance of the solar cells was measured in the ultraviolet–visible–near-infrared region. Quasi-steady-state photoconductance (QSSPC, Sin-ton Instruments) experiments were carried out to measure the minority carrier lifetime of the SiNWs. The SiNW solar cells were also characterized by illuminated current–voltage (I–V) and quantum efficiency measurements. The parameters of a reference solar cell fabricated on the same wafer without any treatment are shown in Table 1.

**Results and Discussion**

The carrier lifetime for the SiNW array without Al₂O₃ could not be measured by QSSPC. Several defects were present on the SiNW surface; these are related to dangling bonds that can lead to a considerable recombination of minority carriers. To passivate the SiNW surface, Al₂O₃ was deposited by ALD, as shown in Fig. 3b, with the Al₂O₃ deposit being embedded into the
SiNW array without space. If there is space in the SiNW/Al₂O₃, this film is easily broken by CMP. Moreover, the lifetime Si wafer with Al₂O₃ increased with the increasing in the thickness of the Al₂O₃ and it tended to be constant from 66 nm as shown in Fig. 4a. From these results, the thickness of the Al₂O₃ layer was set to 66 nm. Figure 4b shows the minority carrier lifetime of each sample as a function of minority carrier density. The minority carrier lifetime of SiNW with Al₂O₃ increased drastically to 65 μs (Fig. 4). Since the dangling bonds were modified by the Al₂O₃, the density of defects decreased. Furthermore improving the minority carrier lifetime of SiNW/Al₂O₃, annealing in forming gas (FG) was conducted and the carrier lifetime was improved to 157 μs. When the carrier lifetime of Si wafer/Al₂O₃ as a function of carrier density was considered, the trend of that with and without annealing are different. In the region of low carrier density, the carrier lifetime increased by the negative fixed charge. On the other hand, the minority carrier lifetime without annealing decreased due to the becoming dominant of Shockley-Read-Hall recombination. Since the negative fixed charge influences the formation of the band bending at the interface between Al₂O₃ and Si surface, the recombination at Si surface can be reduced [25]. We can obtain the information about the existence of negative fixed charge by trend of carrier lifetime as a function of carrier density.

Table 1 Characteristics of a reference solar cell fabricated on the same wafer without any treatment

| Eff (%) | FF  | V_{oc} (V) | I_{sc} (mA/cm²) |
|---------|-----|------------|----------------|
| Ref solar cell | 16.02 | 0.75 | 0.66 | 32.61 |

Therefore, we found that SiNW/Al₂O₃ after annealing was improved by the negative fixed charge. Although SiNWs were completely covered by Al₂O₃, the carriers did not move to the external circuit. Thus, to fabricate the solar cell structure, the Al₂O₃ present on the top of the SiNWs must be removed using an etching paste and applying the CMP technique.

Firstly, an etching paste was used to remove the Al₂O₃ from the top of the SiNW array. After etching, the heterojunction-solar-cell structure was fabricated by forming an n-a-Si/i-a-Si/p-SiNW/i-a-Si/n-a-Si system. Figure 5a shows the I–V characteristics of the SiNW solar cell and the solar cell parameter, series resistance \( R_s \), shunt resistance \( R_{sh} \), ideality factor, and rectification ratios (RR). RR is defined as \( I_F/I_R \), where \( I_F \) (at 0.5 V) and \( I_R \) (at −0.5 V) denote the current at forward and reverse bias respectively. The photovoltaic effect was observed for the SiNW solar cell containing Al₂O₃, and the result shows the removal of Al₂O₃ from the top of the SiNWs. However, the efficiency is low (0.14%) because of the low short-circuit current \( I_{sc} \) and open-circuit voltage \( V_{oc} \) values. In the case of \( V_{oc} \), the carrier lifetimes of SiNWs with Al₂O₃ decreased to 9 μs after using the etching paste. Figure 5b shows a high-magnification top view of the SEM image of a SiNW array with Al₂O₃ after etching. The area in which the SiNWs are exposed is small, and the number of carriers that can be taken out has decreased. Figure 5c shows the low-magnification top view of the SEM image. Since the etching proceeds non-uniformly and the shape before etching was already non-uniform, the non-uniformity of Al₂O₃ increases after etching. We found that it is difficult to
remove the Al₂O₃ uniformly using the etching paste, but to improve the $I_{sc}$ of SiNW solar cells, a uniform etching is required.

CMP was performed to uniformly etch the Al₂O₃ deposited on the SiNWs. Figure 6a and b shows the top-view SEM image of SiNWs with Al₂O₃ after CMP. First, the SiNW array did not break after CMP, indicating that the mechanical strength of the SiNW array with Al₂O₃ is improved by embedding the space between SiNWs. Since CMP can uniformly etch Al₂O₃, the top of the SiNW/Al₂O₃ film became flat.

After CMP, the heterojunction-solar-cell structure was fabricated by forming an n-a-Si/i-a-Si/p-SiNW/i-a-Si/n-a-Si using PECVD system. Figure 7 shows the $I–V$ characteristics of SiNW solar cells with etching thicknesses of 0, 5, and 10 μm and the solar cell parameter, $R_s$, $R_{sh}$, ideality factor, and RR are listed in Table 2. For an etching thickness of 0 μm (when the top of the SiNWs was observed, etching was halted), the photovoltaic effect was confirmed, with a conversion efficiency of 0.8%. $I_{sc}$ of 6.11 mA/cm² was observed. Although the $I_{sc}$ value increased compared with the results obtained for
the etching paste, it is still a low value. The top of the SiNW arrays was aggregated by surface tension in Fig. 4a. Since a part of the SiNWs did not have contact to the a-Si layer, the carriers moved to the external circuit with difficulty. To improve the contact area, the etching thickness was increased to 5 μm, and the $I_{sc}$ increased to 10.3 mA/cm². At an etching thickness of 10 μm, the $I_{sc}$ improved to 14.0 mA/cm². As the aggregated SiNW arrays were removed, the contact area between SiNW and a-Si increased. On the other hand, an extremely low $V_{oc}$ of 0.3 V was obtained. The minority carriers were measured after CMP, and the minority carrier lifetime decreased drastically from 157 to 19 μs because the passivation quality of the Al₂O₃ deposit decreased by CMP. Since the minority carrier lifetime in the region of low minority carrier density declined after CMP, the negative fixed charge decreased. The recombination center on the SiNW surface increased leading to low $V_{oc}$. Furthermore, in the case of wires, the carrier mobility is lowered, because of the scattering of carriers on the surface, and the conductivity is lowered. Although these results indicated that negative fixed charge might be reduced by CMP, the mechanism still is unclear. On the other hand, when the $R_s$, $R_{sh}$, ideality factor, and RR of the etching paste and the CMP result was compared, each parameter of the etching paste is better than that of CMP. Since $R_s$ of CMP is greater than that of etching paste and $R_{sh}$ of CMP is lower than that of etching paste, contamination might remain on the top of SiNW leading to preventing good contact between SiNW and a-Si. Therefore, a further study is required to investigate the improvement of the passivation quality for enhancing the $V_{oc}$ and $I_{sc}$ of solar cells.

The quantum efficiency of 10-μm-long SiNW and c-Si solar cells were compared. In the case of the external quantum efficiency (EQE), the intensity of the SiNW solar cell is mostly lower than that of the c-Si solar cell in Fig. 8a. However, the EQE of SiNW solar cell was improved in the region from 300 to 500 nm. Figure 8b shows the reflectance of the SiNWs and c-Si solar cells,
and it can be observed that the reflectance of the SiNWs device is lower than that of the c-Si one, particularly in the short wavelength region (from 300 to 500 nm) where it is drastically decreased. Although the reflectance of the SiNW solar cell is lower than that of the c-Si solar cell, the EQE of the SiNW device in other regions is lower than that of the c-Si solar cell. Since long wavelength region of light was absorbed in the bottom of SiNWs, the EQE of SiNW solar cell decreased. The internal quantum efficiencies (IQE) of the SiNW and c-Si solar cells were discussed to eliminate the influence of the reflectance. The wavelength region in which the IQE of the SiNW solar cell is higher than that of the c-Si solar cell decreased. In the wavelength region below 340 nm, the IQE of the SiNW device is higher than that of the c-Si solar cell, which results in an increase of the absorption of the SiNWs. The increase of absorption is caused by a light-trapping effect rather than the optical cavity effect. In order to obtain the optical cavity effect using SiNW, the diameter and position of SiNW should be controlled. Since the diameter and position of SiNW fabricated by MAE was random, it is difficult to obtain the optical cavity effect using the SiNW. On the other hand, the random structure of SiNW can have strong light-trapping effect, suggesting that SiNWs fabricated by MAE are promising for crystalline-silicon thinning.

Conclusion
Surface passivation of SiNWs is crucial for their application in solar-cell devices. Al₂O₃ was fabricated by ALD to passivate the dangling bonds. Since ALD can deposit Al₂O₃ over the entire SiNWs, the carrier cannot move to the external circuit. In this study, an etching paste and the CMP technique were applied to etch Al₂O₃ from the top of the SiNWs. With the etching paste, SiNW solar cells with 0.14% efficiency were successfully obtained. However, since the SiNW array was aggregated by surface tension, the contact area between SiNWs and a-Si was small, leading to a low $I_{sc}$. To further improve the efficiency, the etching thickness was increased, and the efficiency could be improved to 1.6% by increasing $I_{sc}$. In the case of the EQE, the intensity of the SiNW solar cell is lower than that of the c-Si solar cell. Since reflectance in short wavelength region from 300 to 500 nm is drastically decreased, the EQE was improved. The IQEs of the SiNW and c-Si solar cells were discussed to eliminate the influence of the reflectance. In the wavelength region below 340 nm, the IQE of the SiNW device is higher than that of the c-Si solar cell, which results in an increase of the absorption of the SiNWs, suggesting that SiNWs are promising for crystalline-silicon thinning.

Table 2 Characteristics of SiNW solar cells with Al₂O₃ removed by CMP

| Etching thickness | Eff (%) | FF | $V_{oc}$ (V) | $I_{sc}$ (mA/cm²) | $R_{s}$ (Ω) | $R_{sh}$ (Ω) | Ideal factor | $I_{F}/I_{R}$ |
|-------------------|--------|----|-------------|------------------|------------|-------------|-------------|-------------|
| 0 μm              | 0.82   | 0.48 | 0.28        | 6.11             | 7.62       | 1693        | 1.54        | 160         |
| 5 μm              | 0.87   | 0.30 | 0.28        | 10.3             | 8.77       | 1647        | 1.72        | 102         |
| 10 μm             | 1.30   | 0.36 | 0.26        | 14.0             | 8.39       | 1654        | 1.65        | 120         |

Fig. 8 a EQE and IQE of a SiNW solar cell and a reference solar cell. b Reflectance of a SiNW solar cell and a reference solar cell
Abbreviations
Al2O3: Aluminum oxide; CMP: Chemical–mechanical polishing; EQE: External quantum efficiency; \( I_{F} \): The current at forward bias; IQE: Internal quantum efficiency; \( I_{R} \): The current at reverse bias; \( I_{SC} \): Short-circuit current; \( I-V \): Current–voltage; RR: Rectification ratios; \( R_s \): Series resistance; \( R_{sh} \): Shunt resistance; SiNW: Silicon nanowire; \( V_{oc} \): Open-circuit voltage

Acknowledgments
Not applicable.

Funding
The work was supported by the Advanced Low Carbon Technology Research and Development Program (ALCA), Japan Science and Technology Agency (JST) and JSPS KAKENHI Grant Number 17 K14921.

Availability of Data and Materials
All data supporting the conclusions of this article are included within the article.

Author’s Contributions
SK carried out the experiment and wrote the initial draft of the manuscript. KG contributed to the sample fabrication and sample measurement. YK supervised the work and finalized the manuscript. TS gave the final approval of the version of the manuscript to be published. All authors read and approved the final manuscript.

Competing Interest
The authors declare that they have no competing interests.

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Received: 6 January 2019 Accepted: 7 March 2019
Published online: 15 March 2019

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