

# Fabrication of silicon nanowire based solar cells using $\text{TiO}_2/\text{Al}_2\text{O}_3$ stack thin films

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## ABSTRACT

*To improve conversion efficiency of silicon nanowire (SiNW) solar cells, it is very important to reduce surface recombination rate on the surface of SiNWs, since SiNWs have large surface area. We tried to cover SiNWs with aluminum oxide ( $\text{Al}_2\text{O}_3$ ) and titanium oxide ( $\text{TiO}_2$ ) by atomic layer deposition (ALD), since  $\text{Al}_2\text{O}_3$  grown by ALD provides an excellent level of surface passivation on silicon wafers and  $\text{TiO}_2$  has higher refractive index than  $\text{Al}_2\text{O}_3$ , leading to the reduction of surface reflectance. The effective minority carrier lifetime in SiNW arrays embedded in a  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer of 122  $\mu\text{sec}$  was obtained, which was comparable to an  $\text{Al}_2\text{O}_3$  single layer. The surface reflectance of SiNW solar cells was drastically decreased below around 5% in all the wavelength range using the  $\text{Al}_2\text{O}_3/\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer. Heterojunction SiNW solar cells with the structure of ITO/p-type hydrogenated amorphous silicon (a-Si:H)/n-type SiNWs embedded in  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  stack layer for passivation/n-type a-Si:H/back electrode was fabricated, and a typical rectifying property and open-circuit voltage of 356 mV were successfully obtained.*

## INTRODUCTION

Silicon nanowires (SiNWs) have recently attracted much attention as one of novel photovoltaic materials. It is expected that SiNW solar cells [1-13] with a relatively thin absorber layer will have the potential to realize sufficient photocurrent [14-22]. It is very important to reduce surface recombination rate on the surface of SiNWs, since SiNWs have large surface area. We confirmed the high passivation effect of  $\text{Al}_2\text{O}_3$  deposited by atomic layer deposition (ALD) on SiNW arrays by measuring the effective minority carrier lifetime [23,24]. Moreover, we fabricated heterojunction SiNW (HJ-SiNW) solar cell structure, and confirmed the solar cell performance [25]. To obtain SiNW solar cells with higher efficiency, it is needed to reduce internal reflection in SiNW solar cells. The previous structure of SiNW solar cells and the reflectance of the fabricated SiNW solar cell are shown in Figs. 1(a, b), respectively. The high reflectance

in the infrared region is due to large refractive index mismatch between the p-type a-Si:H and the n-type SiNWs embedded in  $\text{Al}_2\text{O}_3$ , since refractive index of the SiNW layer was determined by effective medium approximation (EMA) of crystalline silicon and  $\text{Al}_2\text{O}_3$  with low refractive index of 1.6. In this study, to decrease surface recombination, we tried to cover SiNWs with aluminum oxide ( $\text{Al}_2\text{O}_3$ ) and titanium oxide ( $\text{TiO}_2$ ) by atomic layer deposition (ALD), since  $\text{Al}_2\text{O}_3$  grown by ALD provides an excellent level of surface passivation on silicon wafers and  $\text{TiO}_2$  has higher refractive index than  $\text{Al}_2\text{O}_3$ , leading to the reduction of surface reflectance. In addition, optical properties and effective carrier lifetime of the SiNWs with the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer were evaluated.

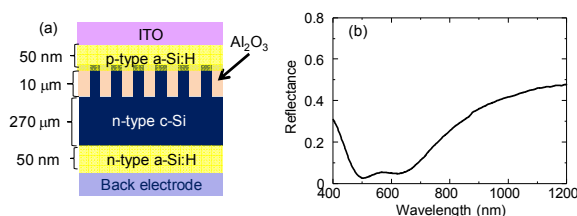


Figure 1. (a) Schematic diagram of previously reported heterojunction SiNW solar cells, (b) Reflectance spectrum of the SiNW solar cell.

## EXPERIMENTAL DETAILS

SiNW arrays were prepared by metal assisted chemical etching with silica nanoparticles (MACES) [26] on a Si wafer (n-type, (100),  $1\text{-}5 \Omega\cdot\text{cm}$ ). Although it is basically same as the MACE process [27], silica nanoparticles are used as an etching mask to control the diameter and density of SiNWs. After the etching of native oxide on a Si wafer, the Si wafer was immersed in ethylenediamine ( $\text{C}_2\text{H}_8\text{N}_2$ ) under nitrogen atmosphere for 3 hours at room temperature. The wafer was rinsed with deionized (DI) water. The hydrophilic Si wafers were immersed in a solution in which 30 nm-silica nanoparticles modified carboxyl group were dispersed at  $2^\circ\text{C}$  for 1 hour. This process formed a dispersed silica nanoparticle layer on the Si wafer. Subsequently a 20 nm-thick silver film was deposited on the wafers with silica nanoparticles using a DC sputtering system. After that, the wafer was chemically etched by using 4.8M HF and 0.15M  $\text{H}_2\text{O}_2$  at room temperature to form SiNW arrays. SiNWs with the length of 1, 5, and 10  $\mu\text{m}$  were prepared on a Si wafer by changing etching duration. The wafers put in  $\text{HNO}_3$  solution to remove silver particles. Finally, the oxide layer existing on the surface of the SiNW array was removed with a HF solution. After the fabrication of SiNW arrays,  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  were deposited by an ALD system (Arradiance, GEM-STAR6). Trimethylaluminum ( $\text{Al}(\text{CH}_3)_3$  (TMA)), tetrakisdimethylaminotitanium ( $\text{Ti}[\text{N}(\text{CH}_3)_2]_4$ ), called TDMAT, and  $\text{H}_2\text{O}$  were used as aluminum, titanium, and oxygen reactant sources, respectively. The TDMAT precursor was heated at  $60^\circ\text{C}$ . Thicknesses of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  were both 20 nm. The other experimental conditions are shown in Table I. After the deposition, post-annealing was carried out at  $400^\circ\text{C}$  to increase negative charge density in the  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$ .

The structure of SiNW arrays was characterized by field emission scanning electron microscopy (FE-SEM) and energy dispersive x-ray spectroscopy (EDS) using JEOL JSM-7001F. The minority carrier lifetime was measured by the  $\mu$ -PCD method using KOBELCO LTE-1510EP. The excited wavelength and intensity of incident light were 904 nm and  $1 \times 10^{14} \text{ cm}^{-2}$ , respectively. The microwave frequency was 9.6 GHz.

Optical properties of the SiNWs were measured by an UV-VIS-NIR spectrophotometer (JASCO V-570) and gas-microphone photoacoustic spectroscopy system. The PAS experimental apparatus consists of a 300 W halogen lamp coupled with a monochromator (Jovin-Yvon H20-IR). The photoacoustic (PA) signal was detected by a microphone and a lock-in amplifier (NF Circuit System 5610B) with a mechanical chopper set at a frequency of about 20 Hz. The wavelength of the excitation light was scanned from 400 to 1600 nm at 5 nm intervals. PA signal intensity was normalized by the signal from a fixed quantity of carbon black to eliminate the effects of the wavelength dependence of the optical apparatus.

A solar cell structure using SiNW arrays covered with  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layers was fabricated as shown in Fig. 2. SiNW arrays with a length of 10  $\mu\text{m}$  were prepared by MACES on a Si substrate. After the fabrication of SiNW arrays, 30-nm-thick  $\text{Al}_2\text{O}_3$ /100-nm-thick  $\text{TiO}_2$ /470-nm-thick  $\text{Al}_2\text{O}_3$  stack layers were deposited on the surface of SiNW arrays by ALD. After the ALD, SiNW arrays were dipped in 1% HF or were etched by reactive ion etching (RIE) with a  $\text{CF}_4/\text{O}_2$  mixture gas to remove the passivation layer on only the top of SiNWs. A p-type hydrogenated amorphous silicon (a-Si:H) layer and an n-type a-Si:H layer were prepared by plasma-enhanced chemical vapor deposition (PECVD) on the front and back, respectively. Indium tin oxide (ITO) was deposited on the p-type a-Si:H layer by RF sputtering. Finally, an Al electrode was evaporated on the back of SiNW solar cells. The final structure was ITO/p-type a-Si:H/n-type SiNW arrays embedded in  $\text{TiO}_2$  and  $\text{Al}_2\text{O}_3$ /n-type a-Si:H/Al. The surface reflectance the SiNW solar cells was evaluated by UV-VIS-NIR spectrophotometers with SHIMADZU Solid Spec-3700. The solar cells were characterized by dark and illuminated  $J-V$  characteristics under AM1.5G illumination.

Table I. Deposition conditions of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$ .

Materials	$\text{Al}_2\text{O}_3$	$\text{TiO}_2$
Temperature	200 °C	225 °C
Cycles	350	144
Flow duration	TMA: 200 msec	TDMAT: 700 msec
	$\text{H}_2\text{O}$ : 200 msec	$\text{H}_2\text{O}$ : 22 msec

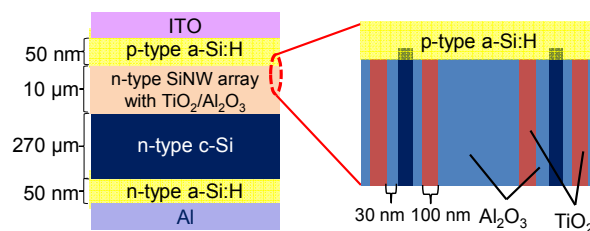


Figure 2. Schematic diagram of the fabricated solar cell structure including SiNW arrays covered with  $\text{Al}_2\text{O}_3/\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layers.

## RESULTS AND DISCUSSION

In Figure 3(a), the cross-sectional SEM images of the SiNW array after the deposition of an  $\text{TiO}_2/\text{Al}_2\text{O}_3$  passivation film are shown. From the figure, the length of SiNWs was  $15.6 \mu\text{m}$ . The dark contrast owing to the gap between SiNWs cannot be confirmed, suggesting that the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack film perfectly covered SiNWs. Figs. 3(b-e) show the EDS mappings of silicon, aluminum, titanium, and oxygen, respectively. White and black signals show a maximum and minimum value, respectively. Compared with Fig. 3(b) and 3(d), the signal intensity contradicts each other, suggesting that the  $\text{Al}_2\text{O}_3$  or  $\text{TiO}_2$  thin films exist between SiNWs. From a SEM image, the shape of SiNWs around the top is needle-like and the gap between SiNWs is about several hundred nanometers. Therefore, the Al and Ti intensities around the top of SiNWs is stronger than the bottom region. These results also suggest that the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack film covered SiNWs from the top to the bottom.

Figure 4 shows external absorption spectra of (a)  $\text{Al}_2\text{O}_3$ , (b)  $\text{TiO}_2$ , and (c)  $\text{TiO}_2/\text{Al}_2\text{O}_3$  coated SiNW arrays. The thickness of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  is 20 nm, respectively. In Fig. 4(a), for the flat substrate, the absorptance in the visible light region decreased mostly due to the high reflectance about 40%. On the other hand, for all the SiNW arrays, almost 100% absorption was obtained, suggesting that fabricated SiNW arrays have very high optical confinement effect. The samples coated with  $\text{TiO}_2$  have the same trend. When both figures were compared, absorptance in near-infrared region, which is generally non-absorption region for bulk silicon, the absorptance spectra between (a)  $\text{Al}_2\text{O}_3$  and (b)  $\text{TiO}_2$  are very different. In the case of  $\text{Al}_2\text{O}_3$ -coated-SiNW, there is few absorption above the wavelength of 1200 nm. This is reasonable because bulk silicon and  $\text{Al}_2\text{O}_3$  do not generally have any absorption in the range except free carrier absorption and absorption related with defects. On the other hand,  $\text{TiO}_2$ -coated-SiNW has very large absorption. Moreover, the absorption was increased with increasing the length of SiNWs. Since the absorption in the region, was not detected in the flat substrate after the deposition of  $\text{TiO}_2$ , it was predicted that absorption sites in the bandgap generated at the  $\text{TiO}_2/\text{SiNW}$  interface. In the case of  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer, although the absorption in the infrared region was reduced a little compared to  $\text{TiO}_2$ -coated SiNW, the high value above 80% was kept. Chen *et al.* have been reported that when lattice disorders in hydrogenated anatase  $\text{TiO}_2$  nanocrystals were introduced, mid-gap electronic states were created, accompanied by a reduced bandgap [28]. According to this paper, it is possible that the disorder may be introduced into  $\text{TiO}_2$  films at the interface between  $\text{TiO}_2/\text{SiNW}$  and the absorption of mid-gap electric states was generated.

Figure 5 shows absorption spectra measured by photoacoustic spectroscopy. Fig. 5 also has the same trend as Fig. 4. The variation of absorption around 1120 nm, which is around the absorption edge of bulk Si, should be noted. In the case of  $\text{Al}_2\text{O}_3$ , the signal was decreased remarkably at the region (Fig. 5(a)). On the other hand, in  $\text{TiO}_2$ -coated-SiNWs, the variation is very small (Fig. 5(b)). This suggests that  $\text{TiO}_2$ -coated-SiNWs have large absorption in longer wavelength than 1120 nm. Such a high absorption in the wide range was also observed in the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack film shown in Fig. 5(c). On the other hand, the flat Si wafer with the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer did not have such a high absorption in the same range, suggesting that both SiNW structure and the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer enhanced the optical confinement. The absorption of an infrared region may be due to the parasitic absorption of disordered  $\text{TiO}_2$ . Although such a parasitic absorption does not contribute to the photoelectric conversion, it is possible to realize efficient photothermal conversion devices [29].

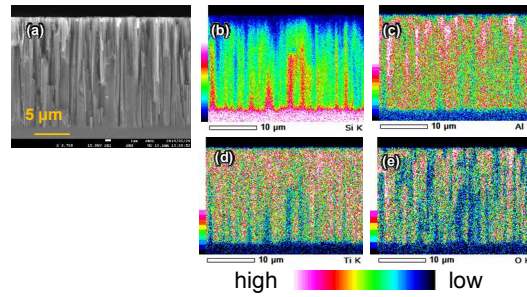


Figure 3 (a) Cross-sectional SEM image and (b-e) EDS mapping of the SiNW array after deposition of  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack film. (b) Si, (c) Al, (d) Ti, and (e) O.

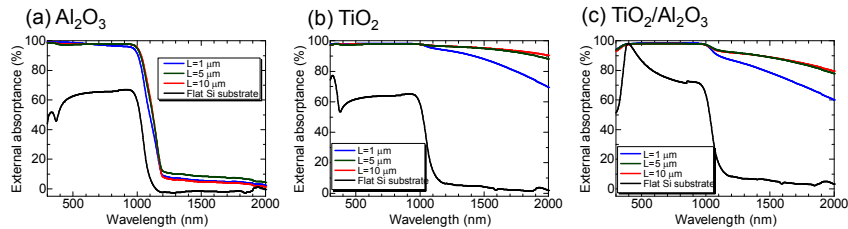


Figure 4. External absorbance calculated from measured transmittance and reflectance.

(a)  $\text{Al}_2\text{O}_3$ , (b)  $\text{TiO}_2$ , and (c)  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack.

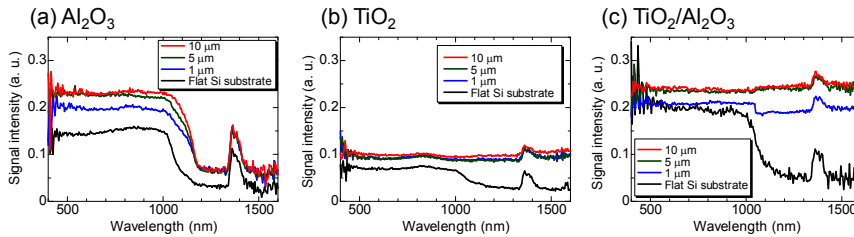


Figure 5 (a) Absorption spectra obtained by PAS.

(a)  $\text{Al}_2\text{O}_3$ , (b)  $\text{TiO}_2$ , and (c)  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack.

The  $\mu$ -PCD measurement revealed that the effective minority carrier lifetime of SiNW arrays embedded in the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer was improved from 6 to 122  $\mu\text{s}$  by the deposition of the passivation layer and post-annealing. It is noted that the effective minority carrier lifetime in  $\text{TiO}_2/\text{Al}_2\text{O}_3$  coated SiNWs was almost same as the  $\text{Al}_2\text{O}_3$  coated SiNWs. It has been reported that since the fine interface between a SiNW and  $\text{Al}_2\text{O}_3$  was formed and dangling bonds on the surface were modified by hydrogen and oxygen, the minority carrier lifetime in the SiNW arrays was improved [23]. Therefore, the improvement was achieved mainly by the passivation effect of  $\text{Al}_2\text{O}_3$ . If the stack layer was applied to the passivation films for SiNW arrays, not only passivation effect

but also lower reflectance could be obtained. It is expected to improve the conversion efficiency of heterojunction SiNW solar cells.

Figure 6(a) shows the reflectance of SiNW solar cells covered with 600-nm-thick  $\text{Al}_2\text{O}_3$  single layer (black solid line), covered with 30-nm-thick  $\text{Al}_2\text{O}_3$ /100-nm-thick  $\text{TiO}_2$ /470-nm-thick  $\text{Al}_2\text{O}_3$  stack layer with HF etching process, and with RIE after the deposition of the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer. From this figure, the reflectance of the SiNW array covered with the  $\text{Al}_2\text{O}_3/\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer with the HF etching process was reduced drastically below 20% compared with SiNW fully covered with  $\text{Al}_2\text{O}_3$ . This is because the refractive index in the SiNW layer was increased. Moreover, in the case of the  $\text{Al}_2\text{O}_3/\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer with RIE, the reflectance was less than around 5% in all the wavelength range. Although the deposition of a passivation layer reduces the surface roughness of the SiNW layer, the RIE process can not only expose the top of SiNWs but also enhance the surface roughness. SiNW arrays fabricated by MACES have a tendency to form bundles after the wet etching process because of the surface tension during drying [17]. From the SEM images, the lateral size of one bundle of SiNWs with the length of 10  $\mu\text{m}$  was about 1–3  $\mu\text{m}$ . This causes Mie-related scattering of light in the wavelength range of visible and infrared light and the optical path length will be increased [30]. Definitely, these results suggest that the usage of  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer and RIE process can reduce the surface reflectance of heterojunction SiNW solar cells drastically.

Finally,  $J$ - $V$  characteristics were measured as shown in Fig. 6(b). Red and blue lines show SiNW solar cells with the HF etching and with the RIE after the deposition of the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer, respectively. In the case of dark  $J$ - $V$  characteristics, a typical rectifying property was successfully obtained for both solar cells, although leakage current can be seen. Compared with two solar cells, better characteristics were obtained in the case of the HF etching. In the case of the RIE, unintentional plasma damage was introduced on the surface of the sample. As a result, minority carrier recombination at the pn interface was increased, leading to the degradation of open-circuit voltage ( $V_{oc}$ ). Moreover, the insufficient etching of  $\text{Al}_2\text{O}_3/\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer increased series resistance ( $R_s$ ) to  $1.14 \times 10^3 \Omega$ , resulting in the decrease of fill factor ( $FF$ ). On the other hand, in the case with the HF etching,  $V_{oc}=356 \text{ mV}$ ,  $J_{sc}=2.96 \text{ mA/cm}^2$ ,  $FF=0.430$ , and  $E_{eff}=0.453\%$  were obtained as shown in Table II. The reason why  $J_{sc}$  did not improved remarkably is that the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer on the top of SiNW was not removed perfectly. Therefore, high  $R_s$  of  $3.51 \times 10^2 \Omega$  was still observed. It is very important to look for the way to remove the  $\text{TiO}_2$  effectively on the top of SiNW arrays. If this issue was resolved, it is expected to improve the conversion efficiency of heterojunction SiNW solar cells due to lower reflectance.

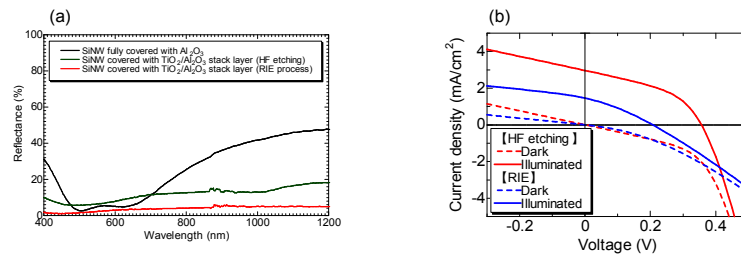


Figure 6. (a) Reflectance of SiNW solar cell covered with 600-nm-thick  $\text{Al}_2\text{O}_3$  single layer (black solid line), covered with 30-nm-thick  $\text{Al}_2\text{O}_3$ /100-nm-thick  $\text{TiO}_2$ /470-nm-thick  $\text{Al}_2\text{O}_3$  stack layer with HF etching process (Green solid line), and with RIE (Red solid line) after the deposition of the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer. (b)  $J$ - $V$  characteristics of SiNW solar cells. Red and blue lines show SiNW solar cells with HF etching and with RIE after the deposition of the  $\text{TiO}_2/\text{Al}_2\text{O}_3$  stack layer, respectively. Dashed and solid lines show dark and illuminated  $J$ - $V$  characteristics, respectively.

**Table II.** Solar cell parameters obtained from the illuminated  $J$ - $V$  characteristics.

Etching method	$V_{oc}$ (mV)	$J_{sc}$ (mA/cm <sup>2</sup> )	$FF$	$R_s$ ( $\square$ )	$R_{sh}$ ( $\square$ )	$Eff.$ (%)
HF	356	2.96	0.430	$3.51 \times 10^2$	$2.94 \times 10^3$	0.453
RIE	207	1.45	0.308	$1.14 \times 10^3$	$2.95 \times 10^3$	0.093

## Conclusions

To improve conversion efficiency of silicon nanowire (SiNW) solar cells, it is very important to reduce surface recombination rate on the surface of SiNWs, since SiNWs have large surface area. To decrease surface recombination, we tried to cover SiNWs with aluminum oxide ( $Al_2O_3$ ) and titanium oxide ( $TiO_2$ ) by atomic layer deposition (ALD), since  $Al_2O_3$  grown by ALD provides an excellent level of surface passivation on silicon wafers and  $TiO_2$  has higher refractive index than  $Al_2O_3$ , leading to the reduction of surface reflectance. The effective minority carrier lifetime in SiNW arrays embedded in a  $TiO_2/Al_2O_3$  stack layer of 122  $\mu$ sec was obtained, which was comparable to an  $Al_2O_3$  single layer. The surface reflectance of SiNW solar cells was drastically decreased below around 5% in all the wavelength range using the  $Al_2O_3/TiO_2/Al_2O_3$  stack layer. The absorptances of SiNWs with the  $TiO_2/Al_2O_3$  stack layer were almost 100% in all the wavelength ranges from 300 to 2000 nm. Although such a parasitic absorption does not contribute to the photoelectric conversion, it is possible to realize efficient photothermal conversion devices. Heterojunction SiNW solar cells with the structure of ITO/p-type hydrogenated amorphous silicon (a-Si:H)/n-type SiNWs embedded in  $Al_2O_3$  and  $TiO_2$  stack layer for passivation/n-type a-Si:H/back electrode was fabricated, and a typical rectifying property and open-circuit voltage of 356 mV was successfully obtained. It is possible to improve the efficiency further by optimizing the conditions of the removal of passivation thin films at the interface between a pn junction.

## Acknowledgments

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