

# Highly efficient industrial large-area black silicon solar cells achieved by surface nanostructured modification



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## ABSTRACT

Traditional black silicon solar cells show relatively low efficiencies due to the high surface recombination occurring at the front surfaces. In this paper, we present a surface modification process to suppress surface recombination and fabricate highly efficient industrial black silicon solar cells. The Ag-nanoparticle-assisted etching is applied to realize front surface nanostructures on silicon wafers in order to reduce the surface reflectance. Through a further tetramethylammonium hydroxide (TMAH) treatment, the carrier recombination at and near the surface is greatly suppressed, due to a lower surface dopant concentration after the surface modification. This modified surface presents a low reflectivity in a range of 350–1100 nm. **Large-area solar cells with an average conversion efficiency of 19.03% are achieved by using the TMAH treatment of 30 s.** This efficiency is 0.18% higher than that of standard silicon solar cells with pyramidal surfaces, and also a remarkable improvement compared with black silicon solar cells without TMAH modifications.

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## 1. Introduction

Surface texturing and antireflection coating on the front surface are known to be very important to reduce the surface reflection and thereby enhance the efficiency of solar cells. Conventionally, a random pyramidal structure formed by wet anisotropic potassium hydroxide (KOH) etching combined with surface antireflection coatings (ARCs) like silicon nitride ( $\text{SiN}_x$ ) based on interference is the industrial method used to reduce the reflection of monocrystalline silicon surface [1,2]. However, the  $\text{SiN}_x$  ARCs deposited on pyramidal structures works only in a narrow spectral range, around 600 nm, and at small angles of incident photons [2–5]. Recently, the black silicon has drawn great attention for nearly ideal photo-absorption across the solar spectrum. The near-zero reflection is obtained by forming the layer with gradually varying refractive index between the air and silicon surface, which is due to the fact that the surface nanostructures are smaller than the incident light wavelength [6–8]. Several methods have been used to fabricate

black silicon, including metal-assisted etching [9,10], laser-induced etching [11], reactive-ion etching [12] and electro-chemical etching [13]. In the past, several authors reported improvements in the conversion efficiency of P-type black silicon solar cells. Oh et al. reported 18.2% efficient silicon solar cells on 0.8081  $\text{cm}^2$  float zone (FZ) material, without any antireflection coating layer [14]. Wang et al. presented a 0.92  $\text{cm}^2$ -large p-type black silicon solar cells passivated with ALD  $\text{Al}_2\text{O}_3$  with an efficiency of 18.2% [15]. Our group has previously developed a 17.3%-efficient 156 × 156  $\text{mm}^2$ -large black silicon solar cells using metal-nanoparticle-assisted chemical etching without any antireflection coating layer [16].

The black silicon solar cell structure has potential for achieving high efficiencies in mass production for nearly ideal photo-absorption across the solar spectrum. However, at present, the efficiency of “black silicon” solar cells is far lower than the commercially available silicon solar cells with the same process except the surface texture method, which is due to increasing surface recombination and Auger recombination as a result of the large surface area and high doping concentration [14,16]. In this paper, the nanotextured front surface, fabricated using Ag nanoparticles after anisotropic KOH etching, is used to reduce the reflectance of silicon surface. The tetramethylammonium hydroxide (TMAH) is added to modify the surface of nanotextured silicon and control the dopant concentration of the emitter. We optimize the process

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for large-area mono-crystalline solar cell with nanotextured surface, and achieved a conversion efficiency up to 19.03% for a large  $156 \times 156 \text{ mm}^2$  mono-crystalline solar cell with nanotextured surface.

## 2. Experimental

As base material, we used  $156 \times 156 \text{ mm}^2$ -large p-type Czochralski (CZ) silicon wafers with resistivity of  $1\text{--}3 \Omega \text{ cm}$  and thickness of  $200 \pm 10 \mu\text{m}$ . After the saw damage removal, all wafers were immersed into aqueous alkaline solutions with KOH (2 wt%) and IPA (2 wt%) at  $80^\circ\text{C}$  for 25 min to form pyramidal structures. After the pyramid-texturing process, the wafers were arranged as back-to-back and immersed into 5 wt% hydrofluoric acid (HF) solution for 30 s and then immersed in deionized (DI) water. Thus, etching resulted in formation of the nanostructure only on the front surface of the wafers. The Ag nanoparticles were electroless-deposited on the silicon surface by immersing the wafer in 0.001 mol/L  $\text{AgNO}_3$  and 1 vol% HF solution for 90 s. Then the wafer covered by Ag nanoparticles was immersed in a mixed solution of 10 vol% HF and 2 vol% hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and DI water for 25 s to form the nanostructure silicon surface. Afterward, the wafers were immersed into the solution containing  $\text{NH}_4\text{OH}:\text{H}_2\text{O}_2:\text{DI}$  in a 1:1:5 volume ratio volume for 5 min to remove Ag nanoparticles. After clean the wafer, phosphorus diffusion was carried out in a standard industrial diffusion furnace with optimized process and the edge isolation etching was made by reactive ion etching. The phosphosilicate glass (PSG) layer, formed during diffusion, was removed by 5 wt% HF solution. Subsequently, a 0.05% TMAH solution was used for treatment for 30 s or 60 s to modify the nanostructures and reduce the surface phosphorus doping concentration. The  $\text{SiN}_x$  coating was deposited by plasma enhanced chemical vapor deposition (PECVD). The front and back metalization of passivated black silicon wafers were carried out using standard screen-printing with Ag-paste and Al-paste respectively, followed by co-firing in a lamp-heated belt furnace. In addition, we also produce some traditional pyramid-textured silicon solar cells with standard production process. The complete cell fabrication process flow is shown in Fig. 1. Finally, we produced four kinds of silicon solar cells: traditional pyramid-textured silicon solar cells as reference (group R), nanotextured solar cells without any modification (group S1), nanotextured solar cells with 30 s

TMAH modification (group S2) and nanotextured solar cells with 60 s TMAH modification (group S3).

The surface morphology of black silicon was analyzed using a Hitachi S-4800 field emission scanning electron microscope (FESEM), and the silicon surface reflection was measured with a Varian Cary 6000i spectrometer using an integrating sphere. The passivated wafers were characterized by quasi-steady-state photoconductance (QSSPC) instrument (WCT-120 by Sinton Consulting) at an injection level of  $\Delta n = 10^{15} \text{ cm}^{-3}$ . The quantum efficiencies and photovoltaic conversion efficiencies of these solar cells were tested by Quality Supervision & Testing Center of Chemical and Physical Power Sources of Ministry of Information Industry.

## 3. Results and discussion

For comparison, the cross-sectional SEM images of the Si wafers subjected to KOH texturing and Ag-assisted etching, and that have modified nanostructures are both presented. Fig. 2(a) is a SEM image of pyramid-textured silicon obtained by etching in alkaline solution. Fig. 2(b) shows the SEM image of nanostructure of black silicon without any modification. It can be seen that the nanopores with a depth of 150–250 nm and a diameter less than 50 nm are formed on the pyramid-textured surface. As shown in Fig. 2(c–d), a majority of the pores of the nanotextured silicon surface modified by 30 s TMAH treatment are about 80 nm in depth and 150–250 nm in diameter, as for the surface modified by 60 s TMAH, the depth of the pores decreased to less than 50 nm and the diameter increased to 300–400 nm. It can be seen that the nanotextured surface area decreases with the TMAH modification because of the reduction in the depth and increase in the pore diameter.

Fig. 3(a) shows the reflectance of pyramid-textured silicon surface, nanotextured silicon surface after 30 s TMAH modification, nanotextured silicon surface after 60 s TMAH modification and nanotextured silicon surface without any modification. The reflectance of the nanotextured silicon wafers are apparently lower than that of the pyramid-textured silicon wafer among the whole wavelength range. The solar-spectrum-weighted average reflectance in the entire wavelength range between 350 and 1100 nm, is 11.3% and 3.9% for pyramid-textured and nanotextured Si wafer, respectively. This is a consequence of the equivalent to a graded index of refraction between air and silicon wafer, when the nanotextured structures are smaller than the wavelength of incident light [6–10]. The average reflectance of nanotextured Si wafer is increased to 6.89% and 9.14% after TMAH treatment for 30 s and 60 s, respectively. It is a high reflectance for the so-called “black silicon”, but it is still much smaller than the reflectance of the pyramid-textured silicon.

Fig. 3(b) shows the reflectance of wafers after the phosphorus diffusion and passivation.  $\text{SiN}_x$  film was still applied as the passivation coating and further ARCs combination with nanostructures on surface, resulting in reduced reflectance. We found that the  $\text{SiN}_x$  film based on quarter-wave-length layers deposited on pyramided silicon wafer perform well only in the wavelength range between 550 and 700 nm ( $<3.5\%$ ) and the average reflectance is 5.33% in the wavelength range between 350 and 1100 nm. The  $\text{SiN}_x$  coating also reduced the reflectance of nanotextured silicon wafers. The average reflectance in the wavelength range between 350 and 1100 nm was 3.78% and decreased to 2.16% after the  $\text{SiN}_x$  film was deposited. The average reflectance of the nanotextured wafer subjected to TMAH modification for 30 s and 60 s was further reduced to 3.51% and 4.58%, respectively. We found that the wafers exhibit a low reflectance because of the combination of the  $\text{SiN}_x$  films and the nanotextured silicon.

The effective carrier lifetime and the emitter saturation current for the wafer were used to characterize the influence of the surface

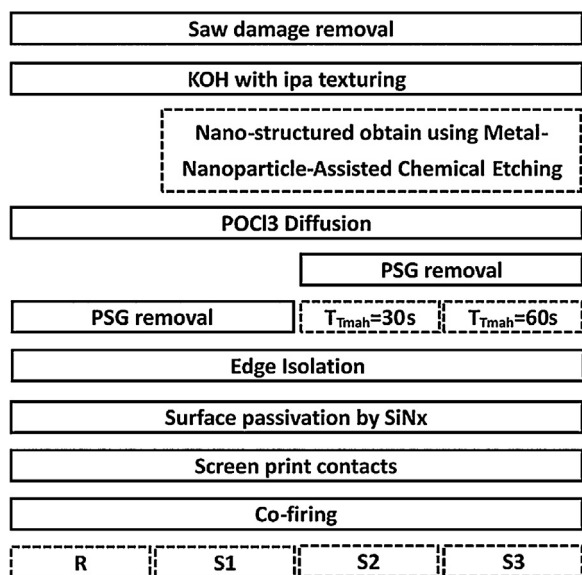
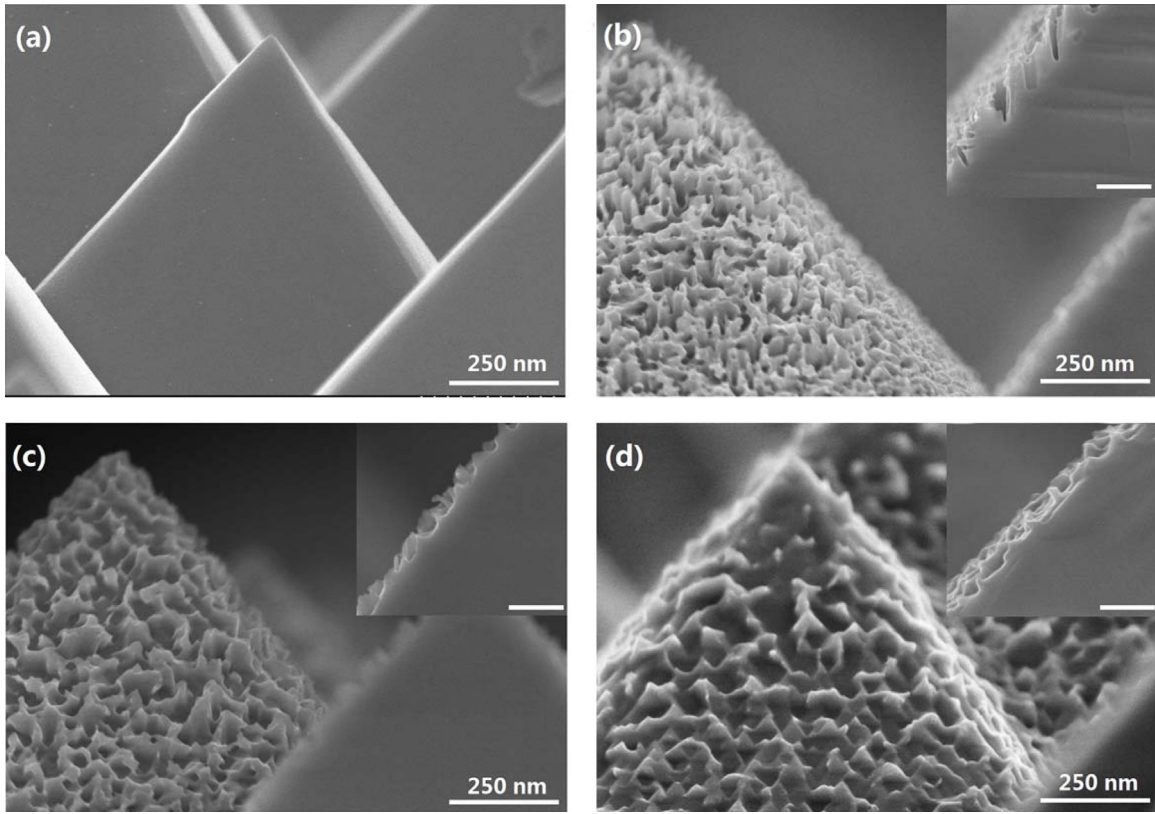


Fig. 1. Processing sequences of our four solar cells presented in Table 1. The dashed boxes highlight the steps that were different in the four process flows.



**Fig. 2.** (a) SEM images of silicon with pyramidal surface. (b) SEM images of silicon with nanotextured surface. (c) SEM images of silicon with nanotextured surface and additionally subjected to TMAH modification for 30 s. (d) SEM images of silicon with nanotextured surface and additionally subjected to TMAH modification for 60 s. The enlarged SEM images of nanopores structures are shown in the insets of b, c, and d (scale bars, 250 nm).

modification with TMAH solution. The test wafers with phosphorus diffusion and  $\text{SiN}_x$  passivation on both sides were utilized in the measurements of the effective carrier lifetime. Here, we introduce an enlargement area factor, defined as  $A_{\text{Nano}}/A_{\text{Micro}}$ , where  $A_{\text{Nano}}$  is the effective area of the nanotextures and  $A_{\text{Micro}}$  is the area of pyramid surface [14]. Fig. 4(a) shows how the effective carrier lifetime increases with the TMAH modification time. At an injection level of  $10^{15} \text{ cm}^{-3}$ , the lifetime of nanotextured wafer was only  $16.74 \mu\text{s}$ . The lifetimes of wafers were increased to  $35.76 \mu\text{s}$ ,  $42.0 \mu\text{s}$ ,  $45.93 \mu\text{s}$  and  $49.83 \mu\text{s}$  after etching in TMAH solution for 15 s, 30 s, 45 s and 60 s, respectively. An significantly increase in the effective carrier lifetime was observed, probably as a result of the decrease in the surface area and the suppression of carrier recombination at the emitter region due to the lower dopant concentration.

The effective lifetime for samples can be expressed as follows [17,18]:

$$\frac{1}{\tau_{\text{eff}}} - \frac{1}{\tau_{\text{Auger}}} = \frac{1}{\tau_{\text{SRH}}} + [J_{0e(\text{front})} + J_{0e(\text{back})}] \frac{(N_A + \Delta n)}{qn_i^2 W} \quad (1)$$

where,  $\tau_{\text{Auger}}$  and  $\tau_{\text{SRH}}$  is the bulk Auger lifetime and bulk Shockley–Read–Hall (SRH) lifetime, respectively.  $N_A$  is the dopant density of the wafer,  $\Delta n$  is the excess carrier concentration,  $n_i$  is the intrinsic carrier density and  $W$  is the wafer thickness.  $J_{0e(\text{front})}$  and  $J_{0e(\text{back})}$  are the saturation current densities of the front and back emitter regions, respectively.

$J_{0e}$  can be calculate by the derivative of the  $(\tau_{\text{eff}} - \tau_{\text{Auger}})$ , provided that the measurement is taken in high injection where  $\tau_{\text{SRH}}$  is constant. In the case of double side nanotextured, diffused and

passivated samples,  $J_{0e} = J_{0e(\text{front})} = J_{0e(\text{back})}$ . So  $J_{0e}$  can be calculate by [19,20]:

$$J_{0e} = \frac{qn_i^2 W}{2} \frac{d}{d\Delta n} \left( \frac{1}{\tau_{\text{eff}}} - \frac{1}{\tau_{\text{Auger}}} \right) \quad (2)$$

Fig. 4(b) shows the saturation current density of nanotextured wafer with and without TMAH modification. It can be seen that the  $J_{0e}$  decrease with the modification time within 60 s. The value of  $J_{0e}$  decreased sharply at modification time 15 s, and then decreased with further increasing the modification time. The  $J_{0e}$  of nanotextured sample was  $754 \text{ fA/cm}^2$ , and then decreased to  $420 \text{ fA/cm}^2$ ,  $322 \text{ fA/cm}^2$ ,  $285 \text{ fA/cm}^2$  and  $270 \text{ fA/cm}^2$  after 15 s, 30 s, 45 s and 60 s TMAH modification. The decrease of  $J_{0e}$  indicates that lower recombination in emitter region or surface.

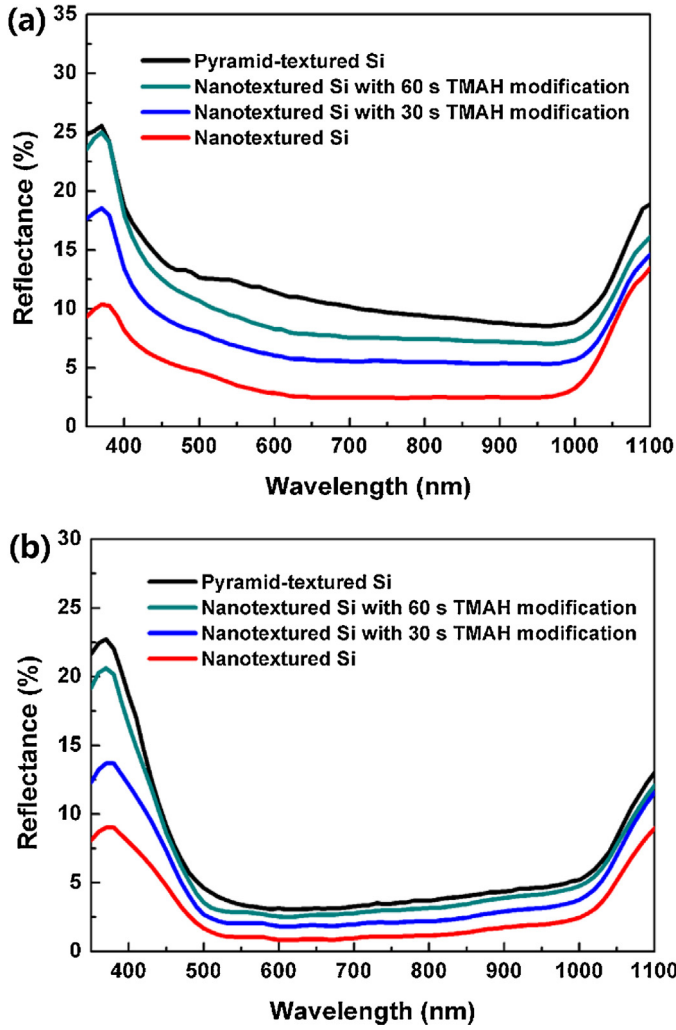
The emitter can be defined as a virtual surface positioned at the edge of the depletion region, and emitter recombination can be considered as a special case of surface recombination ( $S_{\text{eff}}$ ) [20]:

$$S_{\text{eff}} = S_{\text{eff}}^F = S_{\text{eff}}^B \approx J_{0e} \left( \frac{N_A + \Delta n}{qn_i^2} \right) \quad (3)$$

$S_{\text{eff}}$  as function of the modification time is shown in Fig. 5,  $S_{\text{eff}}$  can be counted as follows [14]:

$$S_{\text{eff}} = \frac{A_{\text{Nano}}}{A_{\text{Micro}}} S_{\text{loc}} \quad (4)$$

where  $S_{\text{loc}}$  is the local effective surface recombination velocity, which includes recombination at the surface and the doped region near the surface. If the effective surface recombination velocity is independent of the enlargement in the surface area,  $S_{\text{loc}}$  of samples without TMAH modification and with different modification time will be similar. Fig. 5 shows the  $S_{\text{eff}}$  and  $S_{\text{loc}}$  as function of different

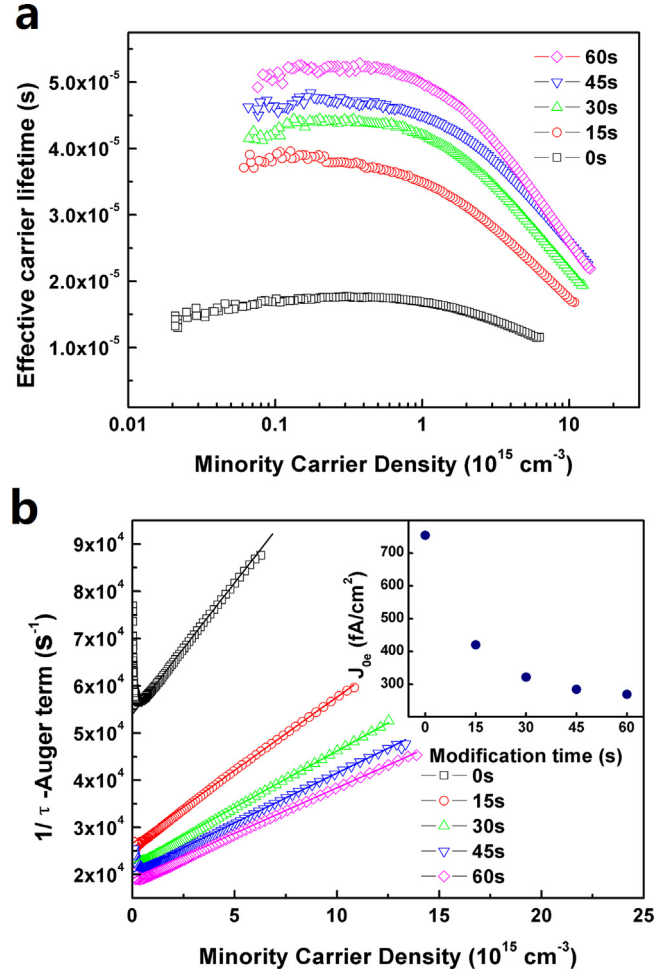


**Fig. 3.** Reflectance vs. wavelength curves: (a) pyramid-textured silicon, nanotextured silicon, nanotextured silicon with 30 s TMAH modification, and nanotextured silicon with 60 s TMAH modification. (b) Passivated by SiN<sub>x</sub> using the substrate sample in (a).

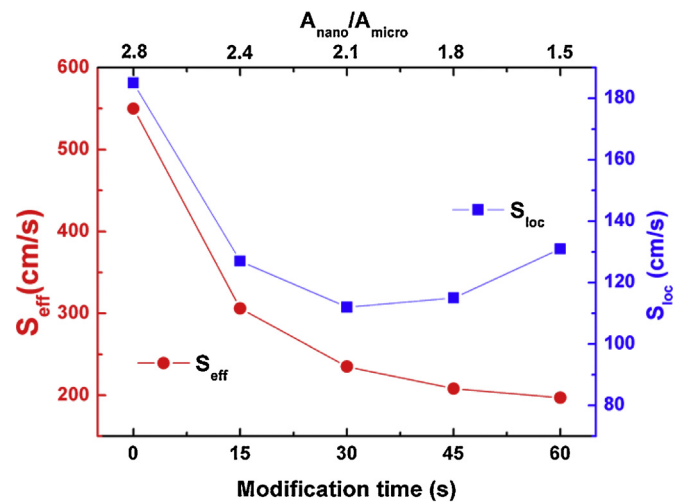
modification time within 60 s. It clearly shows that  $S_{\text{eff}}$  decreases with the increase of modification time within 60 s. The variation of  $S_{\text{eff}}$  as function of modification time reduces as modification time increases.

As shown in Fig. 5, it was found that the value of  $S_{\text{loc}}$  decrease from 185 cm/s to 127 cm/s after 15 s TMAH modification, and then decrease to 112 cm/s after another 15 s. This result confirmed our assumption that the increase of  $\tau_{\text{eff}}$  is due to both of the reduction of surface area and the suppression of Auger recombination in emitter by TMAH modification. The value of  $S_{\text{loc}}$  does not change significantly with the increase in the modification time to 45 s. Our results showed that the optimized modification time is around 30 s, at this time, the P dopant concentration was significantly reduced. The value of  $S_{\text{loc}}$  increased to 132 cm/s with 60 s TMAH modification, which is due to non-uniformity in the surface emitter for the over surface etching.

Table 1 shows the open circuit voltage ( $V_{\text{OC}}$ ), short circuit current density ( $J_{\text{SC}}$ ), fill-factor (FF), and efficiency ( $\eta$ ) of group S1, group S2, group S3 and the group R measured under AM 1.5G. We found that the  $V_{\text{OC}}$ ,  $J_{\text{SC}}$  and  $\eta$  of the group S2 were enhanced 16.35 mV, 1.34 mA/cm<sup>2</sup> and 1.16% (absolute) compared with group S1, though group S2 showed lower reflectance. A high  $V_{\text{OC}}$  was achieved due to (a) the reduced surface recombination owing to



**Fig. 4.** (a) Minority carrier lifetimes and (b) Auger corrected inverse effective lifetime as a function of injection level for nanotextured surface silicon with TMAH modification for 0 s, 15 s, 30 s, 45 s, and 60 s. The samples are presented in Table 1. The solid line is the tangent line at the injection point to determine the saturation current density. The variation of saturation current densities as a function of modification time is shown in the inset of (b).



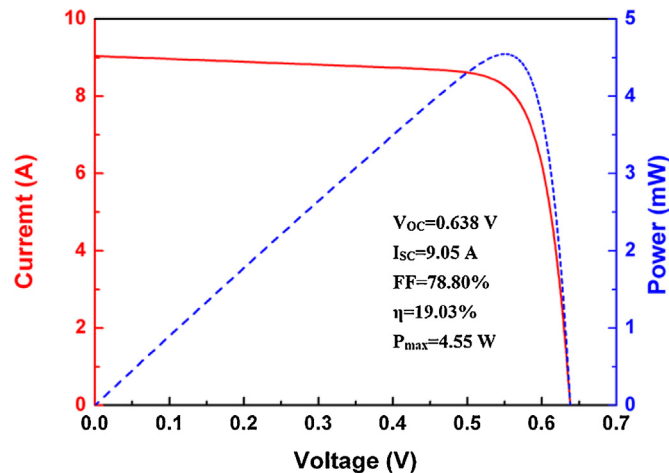
**Fig. 5.** Evaluation of  $S_{\text{eff}}$  (left y-axis) and  $S_{\text{loc}}$  (right y-axis) as a function of TMAH modification time (bottom x-axis) and  $A_{\text{nano}}/A_{\text{micro}}$  (top x-axis).



**Table 1**

Summary of the average electrical parameters of reference solar cells (R), nanotextured solar cells without any modification (S1), nanotextured solar cells with 30 s TMAH modification (S2) and nanotextured solar cells with 60 s TMAH modification (S3). The average values are calculated from 5 to 10 cells.

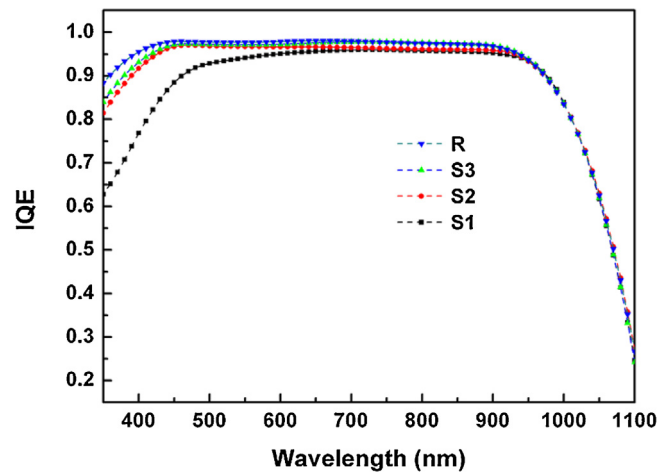
Group	$V_{OC}$ (mV)	$J_{SC}$ (mA/cm <sup>2</sup> )	FF (%)	$\eta$ (%)
S1	620.38 ± 2.27	36.47 ± 0.33	78.85 ± 0.26	17.84 ± 0.12
S2	636.73 ± 0.36	37.81 ± 0.08	78.92 ± 0.16	19.00 ± 0.04
S3	633.93 ± 2.10	37.43 ± 0.19	77.25 ± 0.30	18.33 ± 0.13
R	636.93 ± 1.20	37.36 ± 0.17	79.09 ± 0.13	18.82 ± 0.11



**Fig. 6.**  $I$ - $V$  characteristic of the best nanotextured  $156 \times 156 \text{ mm}^2$  crystalline solar cells.

the decrease in surface area after TMAH modification and (b) the suppression of the Auger recombination in the emitter, which was because of the removal of the “dead layer” using the TMAH solution. This suppression of recombination has positive influence on the passivation of group S2 cells and enhances the collection efficiency of electrons, which results in a higher  $J_{SC}$  and  $\eta$ . However, the  $\eta$  of group S3 decreased to 18.33% due to the slightly decrease in  $V_{OC}$ ,  $J_{SC}$  and the sharply decrease in FF. The decrease of  $J_{SC}$  could be attributed to the reduction of absorption at surface region for the excessively etching time. The low fill factor was caused by the increase in the series resistance owing to the high sheet resistance (over  $120 \Omega/\square$ ). We also prepared an industry-standard mono-crystalline solar cell with pyramid-textured surface, and the electrical performance was presented in Table 1. The  $J_{SC}$  and  $\eta$  of our best black silicon solar cells with TMAH modification are higher than those of the industrial-standard mono-crystalline solar cell, which could be attributed to the lower reflectance of the black silicon solar cells. A nanotextured silicon solar cells has achieved a 19.03% ( $V_{OC} = 0.638 \text{ V}$ ,  $J_{SC} = 9.05 \text{ A}$ ,  $FF = 78.80\%$ ) efficiency on  $156 \times 156 \text{ mm}^2$  crystalline silicon wafer. The  $I$ - $V$  characteristic is shown in Fig. 6.

Fig. 7 shows the IQE of the group S1, S2, S3 and R as a function of wavelength range between 350 and 1100 nm. It also shows an appreciable difference in the wavelength range between 350 and 650 nm between the group S1 and others. The higher dopant concentration in the emitter of group S1, increases the photons absorbed in the emitter and space charge region, which corresponds to an incident wavelength between 350 and 650 nm [14]. The IQE of group S2 and group S3 were improved in the wavelength range between 350 and 650 nm, suggesting that TMAH modification effectively suppress the emitter carrier recombination. Fig. 7 shows a similar IQE between groups S2 and S3, the IQE was slightly enhanced after additional 30 s TMAH modification. Compared to standard pyramid-textured solar cells (group R), the nanotextured



**Fig. 7.** Internal quantum efficiency of reference solar cell, and solar cells that were nanotextured without TMAH modification and that were nanotextured with TMAH modification for 30 s and 60 s.

solar cells subjected to TMAH modification (groups S2 and S3) have lower IQE in the wavelength between 350 and 450 nm, because of the enhanced surface recombination due to the enlarged surface area.

#### 4. Conclusion

Nano-textured front silicon surface and emitter modification were realized using TMAH solution after phosphorus diffusion. Significant improvements in  $V_{OC}$  and  $J_{SC}$  were observed leading to increased solar cell efficiency for modified nanotextured surface compared to the traditional nanotextured surface. We found that the Auger recombination in nanotextured emitters is very high because of the high dopant concentration due to the large bare surface region instead of the pyramid-textured surface in diffusion. The TMAH solution is used to reduce the surface area and suppress the Auger recombination in emitter after phosphorus diffusion. **An increase of 1.16% in cell efficiency was achieved with a gain of 16.34 mV in  $V_{OC}$  and 1.34 mA/cm<sup>2</sup> in  $J_{SC}$ .** This was achieved by the reduction in the Auger recombination in emitter and the decreases in the surface area. Finally, our best solar cells with nanotextured front surface exhibit higher conversion efficiency than traditional pyramid-textured mono-crystalline silicon solar cells. Nano-textured front surface silicon therefore presents great potential for industrial applications.

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