

High-Performance Black Multicrystalline Silicon Solar Cells by a Highly Simplified Metal-Catalyzed Chemical Etching Method

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Abstract—A wet-chemical surface texturing technique, including a two-step metal-catalyzed chemical etching (MCCE) and an extra alkaline treatment, has been proven as an efficient way to fabricate high-efficiency black multicrystalline (mc) silicon solar cells, whereas it is limited by the production capacity and the cost cutting due to the complicated process. Here, we demonstrated that with careful control of the composition in etching solution, low-aspect-ratio bowl-like nanostructures with atomically smooth surfaces could be directly achieved by improved one-step MCCE and with no posttreatment, like alkali solution. The doublet surface texture of implementing this nanobowl structure upon the industrialized acidic-textured surface showed concurrent improvement in optical and electrical properties for realizing 18.23% efficiency mc-Si solar cells (156 mm × 156 mm), which is sufficiently higher than 17.7% of the solely acidic-textured cells in the same batch. The one-step MCCE method demonstrated in this study may provide a cost-effective way to manufacture high-performance mc-Si solar cells for the present photovoltaic industry.

Index Terms—Black silicon (b-Si), light trapping, metal-catalyzed chemical etching (MCCE), multicrystalline silicon (mc-Si), solar cell, surface texturing.

I. INTRODUCTION

THE nanostructuring of silicon surfaces known as black silicon (b-Si) has attracted considerable attention for boosting the efficiency as well as reducing the manufacturing costs of solar cells via sufficient suppression of optical loss at front surface [1]–[7]. In particular, applying nanostructures at the mass production level has very important and practical significance

to multicrystalline silicon (mc-Si) solar cells, which constitute the largest part of the present photovoltaic, but an effective light-trapping technique is still lacking due to the random grain orientations. However, the conversion efficiencies of nanostructured b-Si solar cells are still far from satisfactory, especially for the conventional front-contacted solar cell structure with traditional antireflection coating (ARC) of silicon nitride (SiN_x) [8]–[10]. To date, it is well acceptable that the surface recombination related to the larger surface area of the nanostructures and the Auger recombination at these highly doped nanostructures must be well controlled as low as possible, in order to achieve enhancement in efficiency [11]–[13].

In [9], the influence of the sheet resistance of nanostructures involving the emitter on carrier recombination have been deeply investigated, and the regimes of appreciated doping concentration for an emitter layer to mitigate the Auger recombination have been indicated. For the suppression of surface recombination, a straightforward choice is to design and fabricate novel silicon nanostructures with extremely low surface area but sufficient light harvesting. Our group reported a low-aspect-ratio honeycomb nanobowl structure, which is formed by first oxidizing silicon nanopores and then removing the oxide layer [12]. The enhancement in the surface area of the honeycomb nanobowl is only two times compared to the planar surface, but provides an overall reflection down to 2%, ranging from 400- to 900-nm wavelength. As an industry matching and cost-effective alternative, metal-catalyzed chemical etching (MCCE) was widely used to introduce secondary nanostructures upon the microsize bowl-like surface structures [2], [14]–[17], which were prefabricated by the standard acid texturing in conventional mc-Si solar cell industry. Ye *et al.* reported a novel nanoscale pseudopyramid structure formed by MCCE followed by NaOH modification and shown its effectiveness in improving efficiency [15]. Similar NaOH posttreatments have been utilized by other groups to lower the aspect ratio of the as-fabricated nanopores (caused by MCCE) well below 2 [14], [16], receiving improved blue spectral response and thus enhanced conversion efficiencies over 18% or even higher. Besides the standard acid texturing, the aforementioned b-Si needs several extra procedures: a two-step MCCE to fabricate nanopores, an immersion in dense HNO_3 and buffered HF to dissolve the residual Ag and SiO_2 , an NaOH treatment to lower the surface areas, and several times cleanings by deionized (DI) water to avoid cross contaminations. Obviously, it is a big challenge to successfully integrate so many batch processes into the well-established industrial streamline. Besides, the cost effectiveness and production consistency of

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the b-Si solar cells may also be affected when multiple chemical procedures were introduced. Therefore, although an average improvement of 0.3–0.5% absolute efficiency has been proven by this kind of wet-chemical b-Si technique, simplifying procedures to a large extent is of the most concern before it can be broadly accepted by photovoltaic industry.

In this paper, an improved technique with one-step MCCE processing and without alkaline posttreatment is demonstrated for fabricating the black mc-Si. The well-designed reaction solution contains an ultralow amount of AgNO_3 (i.e., the one-tenth of the conventional MCCE), which can directly introduce uniform and shallow nanobowl-like structures upon the microbowl-like structures that were prefabricated via standard acid texturing on mc-Si surface. Our one-step MCCE process also has a polishing effect that offers atomically smooth surfaces to the nanobowl structures with potentially minimized defect density. In comparison with the standard acidic-textured case, the nanobowl/microbowl dual structures show not only improved light-trapping properties but enhanced minority carrier lifetime as well. As a result, the mc-Si solar cells ($156 \text{ mm} \times 156 \text{ mm}$ in area) fabricated by our improved MCCE technique received an average conversion efficiency (η) of 18.09% and short-circuit current (I_{sc}) of 8.765 A, respectively, with 2.3% and 2.4% higher than that of reference cells.

II. METHOD FOR EXPERIMENT

All experiments were performed on p-type mc-Si wafers with resistivity, thickness, and size of around $2 \Omega\cdot\text{cm}$, $180 \mu\text{m}$, and $156 \times 156 \text{ mm}^2$, respectively. For fabricating the nanobowl/microbowl dual-structure textured solar cells, the wafers were first immersed into the standard acidic solution (HNO_3 : $\text{HF} = 2:1$, at 6°C) to form the microscale structures and were then dipped into $\text{AgNO}_3/\text{HF}/\text{H}_2\text{O}_2$ mixed solution (AgNO_3 below $0.0001 \text{ mol}\cdot\text{L}^{-1}$, HF in $0.05\text{--}5 \text{ mol}\cdot\text{L}^{-1}$, and H_2O_2 in $1\text{--}10 \text{ mol}\cdot\text{L}^{-1}$) at room temperature for 1 min to create the nanoscale textures. It is worth noting that the consumption of AgNO_3 here is only about one tenth over the previously reported two-step MCCE process. After that, the wafer was immersed into dense HNO_3 solution for 3 min and then rinsed by DI water to remove the residual Ag nanoparticles. After the texturing process, phosphorous diffusion using POCl_3 was performed at 810°C for 40 min to form the n-type emitters with a sheet resistance of around $110 \Omega/\text{sq}$. Standard rear-junction isolation and phosphorus silicon glass removal were subsequently performed in an industrial inline wet-chemical tool. With the aims of ARC and passivation, a stack of double-layer SiN_x film was deposited at 477°C using an inline direct plasma-enhanced chemical vapor deposition (PECVD) system, consisting of a 20-nm-thick bottom SiN_x layer with a refractive index of 2.25 and a 60-nm-thick top SiN_x layer with a refractive index of 1.95. Finally, the Ag front electrodes and Al back-surface contacts were formed by screen printing and cofiring in a lamp-heated belt furnace system. For comparison purpose, the solar cells based on the pure acidic texture and the acidic texture combined with previously reported nanopores structures by two-step MCCE [18]–[20] were also fabricated in the same batch.

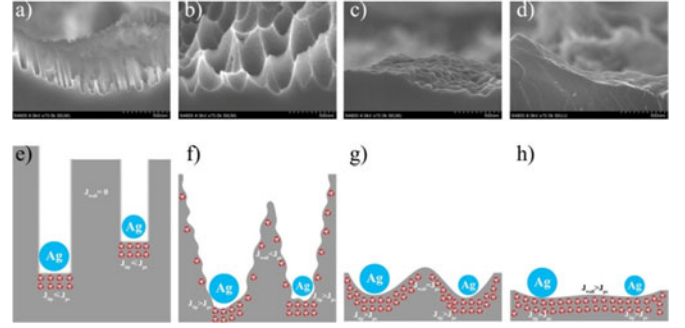


Fig. 1. (a)–(d) Cross-sectional SEM images and (e)–(h) schematics of the mechanism proposed for the formation of mc-Si samples after MCCE for $\rho = 25, 20, 15$, and 7 , respectively.

The textured structures of mc-Si wafers were analyzed by field emission scanning electron microscope (Hitachi S-4800). The crystal structures were determined by imaging with a high-resolution transmission electron microscope (HR-TEM, Tecnai F20). The surface reflections were examined by spectrophotometer (Helios LAB-rc) using an integrating sphere in the wavelength range of $375\text{--}1075 \text{ nm}$. The electrical characteristics of the mc-Si solar cells were investigated by photocurrent–voltage (I – V) measurement under the illumination of AM 1.5G using a solar-simulated light source. The external quantum efficiency (EQE) measurement in the wavelength of $300\text{--}1075 \text{ nm}$ using a 300-W xenon lamp with a spot size of $1 \times 3 \text{ mm}^2$ was calibrated with a silicon photodetector from Newport. The minority carrier lifetime was measured by microwave photoconductivity decay (WT-2000 PVN, Semilab).

III. RESULTS AND DISCUSSION

In a typical one-step MCCE process, the deposition of metal catalysts and the dissolution of silicon are simultaneously happened in the mixed solution that contains H_2O_2 , AgNO_3 , and HF . As shown in reaction (1), the introduction of AgNO_3 gives rise to the generation of Ag on the silicon surface. The dense Ag nanoparticles (Ag NPs) could accelerate holes injection with the presence of oxidizing agent (H_2O_2) and heavily shorten the diffusion distance of the injected holes in the vicinity of Ag NPs [1], [21], [22], as shown in reaction (2). Finally, the nanostructures are formed by the hole-induced silicon oxidation and the HF-driven dissolution of silicon oxides, referring to reaction (3). In brief, the final nanostructure configurations are mainly controlled by the density of the injected holes and their diffusion length, which are directly related to the ratios of the H_2O_2 and HF once the amount of AgNO_3 is fixed

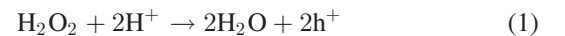


Fig. 1(a)–(d) showed the cross-sectional SEM images of the nanostructures formed on the acidic-textured Si surface by our one-step MCCE with different ρ , where ρ represented the molar ratio of $[\text{HF}]/([\text{HF}] + [\text{H}_2\text{O}_2])$. From Fig. 1(a)–(d), formation

of nanopores, enlarged craters, polished shallow nanobowls, and even flat surfaces are observed as ρ progressively decreases from 25%, 20%, and 15% to 7%. This phenomenon clearly showed that the decreasing proportion of HF induced both the enlargement of the pore diameter and the formation of nearly cone-shaped pores with Ag nanoparticles at the bottom, whereas the increasing in H_2O_2 amount led to the rising of the opening angle of the craters [23]. The one-step MCCE process is actually known to be a mixed electroless and a chemical process. This electrochemical step implies that there are anode and cathode sites on the silicon surface with local cell currents that originate from holes injected in silicon by H_2O_2 . In particular, the H_2O_2 concentration can be considered as the equivalent of this current density in electrochemistry, while the HF concentration rules the surface chemistry and the dissolution of silicon oxides. For the convenience of discussion, the critical current density (J_{ps}) was introduced, below or above which porous silicon formation or polishing occurs. The formation of structures for varying ρ can, thus, be schematically demonstrated in Fig. 1(e)–(h), respectively. For the case of $\rho = 25\%$, the current density was well below J_{ps} ($J_{\text{tip}} < J_{\text{ps}}$), Ag nanoparticles dig pores into bulk Si, and the hole injection was localized at the Si/metal nanoparticle interface due to the catalytic properties of the metal. Thus, the Si was etched into porous Si, as shown in Fig. 1(a). With the decreasing of ρ ($\rho = 20\%$), where the etching current was higher than critical current density ($J_{\text{tip}} > J_{\text{ps}}$), an oxide layer was built at the Si/Ag interface due to the large H_2O_2 concentrations. Then, the injected holes were diffusing from the pore tip to pore walls as a spread current, which is lower than J_{ps} ($J_{\text{walls}} < J_{\text{ps}}$). Hence, the dissolution was isotropic and resulted in a polished surface at the pore tip and microporous Si formed on the pore walls, as shown in Fig. 1(b). While further decreasing ρ down to 15%, the etching current was still higher than the critical current density ($J_{\text{tip}} > J_{\text{ps}}$), but the spread current may very close to J_{ps} ($J_{\text{walls}} \approx J_{\text{ps}}$), in this case, a mixed regime of polishing and porous Si formation led to a polished surface not only at the pore tip but on the pore walls as well. For this reason, the etching surface was becoming much smoother [see Fig. 1(c)], causing a surface texture quite similar to the alkali treatment. Finally, when both the etching current and the spread current were sufficiently higher than the critical current density ($J_{\text{tip}} > J_{\text{ps}}$, $J_{\text{wall}} > J_{\text{ps}}$, for $\rho = 7\%$), where the oxidized layer thickened and covered the entire surface. Thus, the dissolution was isotropic, independent of metal nanoparticles location and resulted in a polished surface, as shown in Fig. 1(d).

In order to evaluate the light-harvesting characteristics of the aforementioned four structures, the reflectance in the wavelength range of 375–1075 nm was measured, as shown in Fig. 2(a). In contrast with the pure acidic textures, the as-fabricated hierarchical structures effectively reduced the reflectance over the entire band regions and exhibited the reduced average reflectance from 31.93%, 16.80%, 10.37%, and 4.85%, for the structures of $\rho = 7\%$, 15%, 20%, and 25%, respectively. The superior light-harvesting property of the hierarchical structures that fabricate under $\rho = 7\%$, 15%, 20%, and 25% could be attributed to two effects: the scattering effect of the microbowl-like structures (acidic texture) with a feature size of 2–6 μm

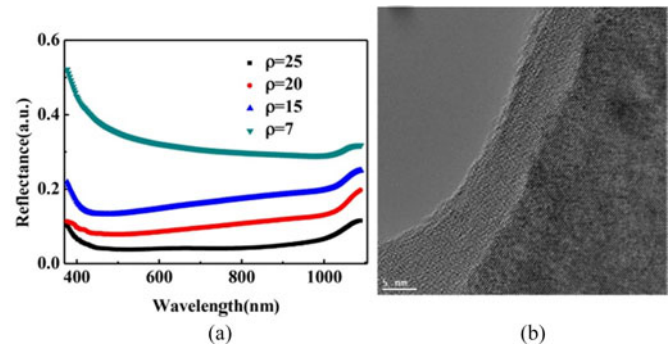


Fig. 2. (a) Reflection spectra of mc-Si samples after MCCE for $\rho = 25, 20, 15$, and 7 in the wavelength range of 375–1075 nm. (b) HR-TEM image collected at a local surface of the nanobowls.

and the gradient refractive index effect of the nanostructures (MCCE texture) with lateral sizes ranging from 30 to 200 nm. It was worth noting that the additive nanobowl-like structures with depth well below 50 nm [see Fig. 1(c)] could help to lower average reflectance of the acidic texture from 31.93% to 16.80%. This value is low enough because the subsequent ARC has not been taken into account. The one-step MCCE with ρ around 15% was, therefore, selected as the preferential recipe to deliver nanobowl/microbowl dual structures for the potential advantages of increased light harvesting and mitigated surface recombination loss. To further investigate the surface quality, an HR-TEM image was randomly collected at the local surface of the nanobowls, as shown in Fig. 2(b). The HR-TEM image was done after 3-min HNO_3 treatment. The HR-TEM image representatively showed that the surface was atomically smooth and the crystalline structure maintained perfect instead of the quite rough surfaces in previously reported MCCE processes, suggesting that the Ag-related MCCE process at the condition of $\rho = 15\%$ does have a strong polishing effect on the Si surface. The surface defect density of the nanobowls is, thus, considered to be highly suppressed.

As well known, it is still a big challenge for industry to achieve a good passivation on nanotextured Si surface utilizing conventional PECVD- SiN_x because the uniform SiN_x layer can rarely be conformably coated on the structures with high aspect ratio. Fortunately, this obstacle does not exist in our case. Fig. 3(a)–(c) showed the SEM images of acidic texture, acidic texture with conventional two-step MCCE texture (conventional black-Si structure), and acidic texture with our nanobowl/microbowl dual structure, respectively, all coated with an 80-nm SiN_x layer. One could clearly see that the uniform SiN_x layer can only be conformably coated on both the surfaces of the acidic texture [see Fig. 3(a)] and the nanobowl/microbowl dual structure [see Fig. 3(c)], but not the conventional black-Si structure. Additionally, the effective minority carrier lifetime (τ_{eff}) measurement using microwave photoconductivity decay was employed to evaluate the surface/interface defects, which greatly influenced the photovoltaic characteristics. The scanned lifetime images on total area ($156 \times 156 \text{ mm}^2$) of the three samples with only front-sided SiN_x coating were shown in Fig. 3(d)–(f), respectively. Although the samples used for characterizations of

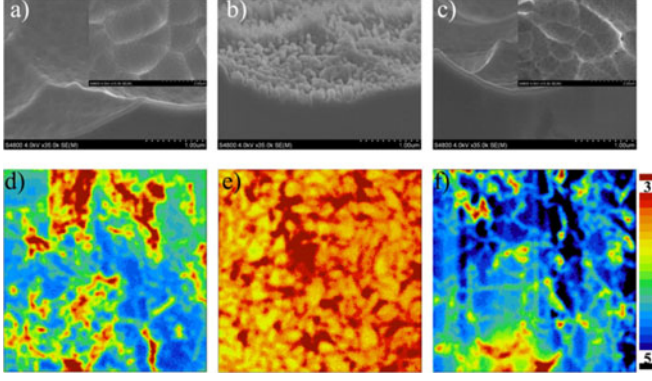


Fig. 3. (a)–(c) SEM images and (d)–(f) minority carrier lifetime mapping of standard acidic texture, conventional black-Si structure, and nanobowl/microbowl dual structure after coated with SiN_x . The insets in (a) and (c) are top-viewed SEM images.

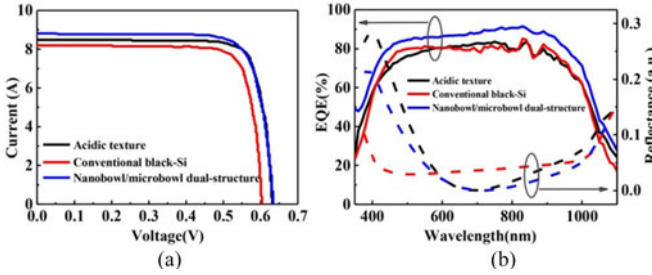


Fig. 4. (a) and (b) Current–voltage (I – V), EQE characteristics, and reflection spectra of three mc-Si solar cells with standard acidic texture, conventional b-Si, and acidic texture with our nanobowl/microbowl dual structure.

TABLE I
PHOTOVOLTAIC PROPERTIES OF THE THREE TYPES OF MC-SI SOLAR CELLS

	V_{oc} (V)	I_{sc} (A)	FF	E_{ff} (%)
Acidic texture	0.631	8.481	0.804	17.70
b-Si	0.603	8.188	0.795	16.15
Micro/nano dual structure	0.633	8.817	0.794	18.23

τ_{eff} were not strictly symmetrical configuration with double-sided coatings, the surface recombination properties can still be indirectly evaluated by measuring the τ_{eff} , while the thickness of device was constant. The averaged τ_{eff} for the samples of acidic texture, conventional black-Si structure, and the nanobowl/microbowl samples were 4.018, 3.287, and 4.495 μs , respectively. Our nanobowl/microbowl dual structure showed the highest τ_{eff} with the best uniformity even in comparison with the standard acidic texture. This could be attributed to the effective reduction of the surface defects and the creation of atomically smooth Si surfaces, which was also supported by TEM observation.

All structures were fabricated into solar cells with the standard alumina back-surface-field configuration, and the current–voltage (I – V) curves and EQE spectra were shown in Fig. 4. Photovoltaic characteristics were summarized in Table I, while statistical parameters were shown in Fig. 5.

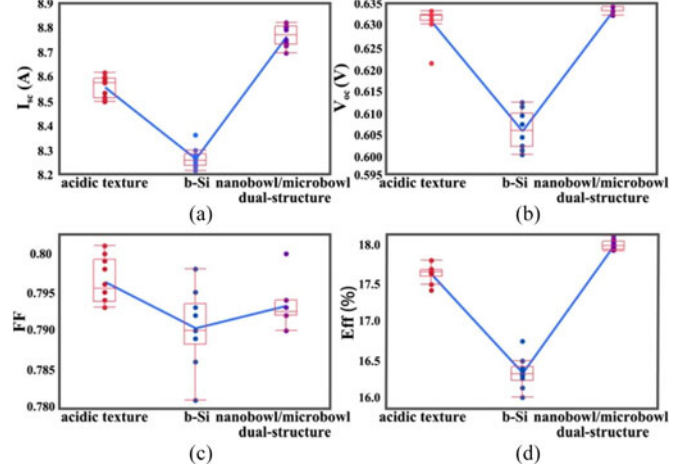


Fig. 5. Electrical parameters statistically collected from a large quantities of mc-Si solar cells with the three different surface textures in a standard production line.

It was evident that the conversion efficiency (η) and the short-circuit current (I_{sc}) of solar cell fabricated by our one-step MCCE method were much higher than the other two kinds of samples, while the open-circuit voltage (V_{oc}) keeps identical. The medium fill factor (FF) of our novel one-step MCCE sample might attribute to the contaminations originated from the processing. As shown in Fig. 4(b), the poor I_{sc} of the two-step MCCE texture stemmed from the low quality of passivation, as well as the deteriorated light-harvesting properties at the wavelengths beyond 600 nm that was caused by the ARC coating [see Fig. 3(b)], whereas its deep nanopores would lead to severe leakage problem and, thus, result in poor V_{oc} and FF . The better EQE performance in the whole wavelength range of our improved one-step MCCE sample further demonstrated its superiorities in light trapping and surface passivation, echoing with the pronounced improved η of 18.23%. The inconsistency between the curves of EQE and reflection for conventional black-Si and our nanobowl/microbowl dual structures at short- and long-wavelength regions can be ascribed to the serious surface recombination arising from the greatly enlarged surface area at front and rear surface, as well as the poor front SiN_x coating, due to the deep and dense nanostructures on both sides of the black-Si that were formed during the MCCE process. Compared with the solar cells with conventional acidic texture, the cells with nanobowl/microbowl structures showed average enhancements in V_{oc} , I_{sc} , and η of 0.4%, 2.4%, and 2.3%, respectively.

IV. CONCLUSION

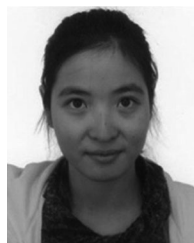
In summary, we have demonstrated an improved MCCE method for fabricating black mc-Si solar cells with efficiency up to 18.23%. The novel nanobowl/microbowl surface structures enabled excellent antireflection, polished surfaces and conformal coating of SiN_x , giving rise to a concurrent improvement in electrical and optical characteristics of the solar cells. As a result, the novel MCCE textured mc-Si solar cells had about a 336 $\text{mA}\cdot\text{cell}^{-1}$ increase in the short-circuit current density and no decrease in open-circuit voltage compared with the regular

acidic textured mc-Si solar cell. More importantly, the highly simplified MCCE technology might provide huge cost effectiveness in manufacturing high-performance mc-Si for the photovoltaic industry.

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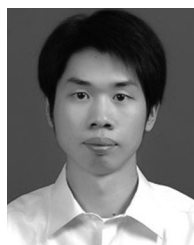
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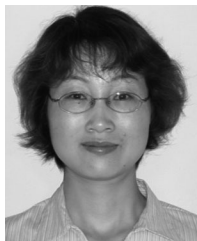
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