Improved Surface Passivation by Wet Texturing, Ozone-Based Cleaning, and Plasma-Enhanced Chemical Vapor Deposition Processes for High-Efficiency Silicon Heterojunction Solar Cells

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Silicon heterojunction (SHJ) solar cells rely on excellent surface passivation of the crystalline wafer. This article reports on the development of wet chemical processes varying the texturing and optimizations of the final clean processes for Czochralski–silicon wafers used in SHJ solar cells. Three different additives are used to modify both the pyramid size and the texture homogeneity on the wafers. As an alternative to standard Radio Corporation of America (RCA) sequence, ozonized deionized (DI) water-based procedures are used to clean the silicon surfaces, reducing process time and the amount of chemicals to obtain the same cleaning quality. In addition, two different amorphous silicon (a-Si:H) passivation stacks are discussed in this article demonstrating an improvement in the open-circuit voltage, $V_{oc}$, by 15 mV. Combining these wet chemistry improvements with optimized process conditions for the passivating a-Si:H layers deposited by plasma-enhanced chemical vapor deposition (PECVD), allows to obtain high-efficiency devices both in small- and full-area solar cells. On 6 in. full-area solar cells (215.3 cm$^2$ aperture area), open-circuit voltages, fill factors, and efficiency values of 738 mV, 81.4%, and 23.2% are obtained, respectively; for 4 cm$^2$ cell size, the best obtained values in equivalent devices are 741 mV, 80.6%, and 23.2%.

1. Introduction

Silicon heterojunction (SHJ) solar cells have been of great interest to the community in the last decades due to their high-efficiency potential.\cite{1} Currently, the highest power conversion efficiency has been demonstrated by Kaneka, with 25.1% in a two-side contacted structure\cite{2} and 26.6% in an all-rear-contacted structure.\cite{3} Despite the efficiency advantage of the all-rear-contacted devices, improvements in the two-side contacted solar cells are still of great interest due to its lean fabrication process with only four main process steps each on both sides of the wafer: 1) wet chemistry to texture and clean the silicon wafers; 2) plasma-enhanced chemical vapor deposition (PECVD) of amorphous silicon (intrinsic and doped); 3) sputtering of transparent conductive oxides (TCOs); and 4) screen printing of silver metal grids.

Passivation of the silicon wafer surfaces is a very important topic independent of the silicon-based technology (a-Si/c-Si heterojunction, thermal diffusion, TopCon, or POLO), and it has been studied extensively in the past.\cite{4-6} To improve the passivation of the SHJ solar cells, several techniques need to be applied: homogeneous wet texturing and/or wet chemical procedures to clean the wafer surfaces; surface passivation by means of intrinsic amorphous silicon (i-a-Si) layers to reduce the recombination losses at the heterointerface; deposition of doped layers to obtain additional field-effect passivation; and gentle TCO and metal-grid-deposition techniques. This article focuses on the additive variation in the solutions used for wet texturing of silicon wafers, on wafer cleaning procedures and on the surface passivation by means of a-Si:H layers deposited by PECVD. The additive used during the wet texturing process determines the pyramid size and structure of the silicon surfaces. In this study, three different additives were tested to compare their influence on the performance of full-area SHJ solar cells. In addition, an ozone-based bath is compared with the standard Radio Corporation of America (RCA) cleaning process, aiming at the reduction of chemical waste without compromising the
quality of the surface cleaning. Finally, this article will discuss how to further improve the passivation of the SHJ solar cells using a new i-a-Si layer stack.

2. Experimental Section

The SHJ solar cells in this study were fabricated on both 5 and 6 in. (M2) n-type CZ wafers with resistivities of approximately 5 and 2 Ω cm and wafer thicknesses of 125 and 150 μm, respectively. All wafers were wet chemically etched in KOH to remove the saw damage, subsequently textured and cleaned. Studies on the texturing process were already done in the past by different groups improving the reproducibility and increasing the throughput in industrial production lines using the same chemical bath thanks to the use of a new additive to the solution.\(^7\) These additives have the function to remove the hydrogen bubbles from the wafer surface, to enhance the pyramid formation improving its wettability.\(^8\) The wet chemical experiments done in this work compare three different additives used in the KOH texturing bath: a reference additive initially used as the standard process,\(^9\) and additives 1 and 2, as the alternative ones provided by Innovative Chemie für Industrie und Umwelt in Berlin GmbH (ICB). A variation of the final cleaning step was also done to implement a process with a smaller chemical consumption than the standard RCA.\(^10\) An ozone bath was used as a more environmentally friendly and lower cost alternative to the RCA. After the final wet chemical clean, a 1% HF dip was performed to remove the native oxide prior to the intrinsic and doped amorphous (a-) and nanocrystalline (nc-) silicon layers deposited by PECVD using an Applied Materials AKT1600 cluster tool. Further details regarding the PECVD layers can be found elsewhere.\(^11\) Minority carrier lifetime measurements were done after the PECVD depositions with a Sinton Instruments WCT-120 instrument on the cell precursors and on symmetric test samples. The contacts of the SHJ solar cells consist of a 75 nm thick indium tin oxide (ITO) layers on both sides of the wafer, as TCO, and a low curing temperature screen-printed silver grid. Figure 1a shows the cross section of the SHJ solar cells. To define the solar cell area, aligned shadow masks were used on both sides of the wafer during sputtering to define 14 cells with an area of 4 cm\(^2\). The ITO on the full-area solar cells was deposited without any mask on the illuminated (front) side and with a shadow frame covering 2 mm of the edges on the rear side. The screen-printed grids on small-area (4 cm\(^2\)) solar cells have a busbar-frame outside the active area, whereas the full-area cells can have two layouts, one with five busbars and another one busbar-less. More details about the SHJ solar cell processing and optimization can be found elsewhere.\(^12,13\) The completed small-area devices were tested with a dual-source Wacom WXS-155S-L2 AAA+ solar simulator under standard conditions (AM1.5G, 25 °C), whereas the full-area devices were tested using a WaveLabs Sinus-220 AM1.5G LED sun simulator. The five busbar full-area devices were measured with an in-house made five bars system, whereas the busbar-less devices were measured using a PCB-Touch device from Meyer Burger (former Pasan).

![Figure 1. a) Schematic cross section of the rear-junction, bifacial SHJ solar cells used in this work. b) FF versus \(V_{oc}\) of SHJ full-area solar cells processed using three different additives, in the inset a picture of the full-area SHJ solar cells shows the five busbar metal grid structure used in this experiment. SEM images of three silicon wafers textured with the three different additives are shown in c) reference HZB, d) additive 1, and e) additive 2. The \(J_{sc}\) values of the best cells for each variation are shown in the same SEM images.](image-url)
To test the passivation homogeneity on the full wafer, photoluminescence images were taken on the full-area SHJ solar cells with five busbars from the texturing experiment using a homemade system with a software and Si-CCD detector from Greateyes. The texturing was also investigated with scanning electron microscopy (SEM) images performed with a HITACHI S-4100 tool. Optical images of the full 6 in. wafers were done with a high-resolution scanner (Epson Perfection V39, scan resolution set to 1200 dpi) on as-textured wafers with different texturing solutions to observe the improvement in homogeneity of the texturing process.

### 3. Results

#### 3.1. Wet Texturing Treatments

Homogenously textured wafer surfaces are crucial for full-area size solar cells performance. To obtain uniformly textured surfaces with optimized pyramids size, the use of the right additive is of great importance. In this article, three different additives were tested to improve the passivation and the homogeneity of the textured wafers. The first additive is a reference, which has been used in the standard process. As an alternative, CellTex ULTRA additives were provided by ICB; additive 1 and additive 2 are two versions of this additive, which were used to improve the wettability of the silicon surface, improving the homogeneity of the texturing. Figure 1b shows the fill factor (FF) versus the open circuit voltage ($V_{oc}$) of differently textured full-area SHJ solar cells with a five busbar layout (inset). Despite the small statistics in the experiment, it is clear that both alternative additives provide a more uniform surface, as shown in Figure 2a,b, which directly transfers to improved solar cell electrical parameters: FF and $V_{oc}$ on the cells processed with the alternative additives are ≈2% rel. and ≈3 mV higher, respectively, compared with the reference cells. In addition, in Figure 1c–e, SEM images of the different wafers show the varying pyramid structures obtained with the different additives. Although the pyramid size of the textured wafers varies when using the different additives, 3–5 μm with additive 1 (Figure 1d) and 1–3 μm with the reference and additive 2 (Figure 1c,e), the short-circuit current density ($J_{sc}$) of the solar cells is in the same range on all samples (≈35.6 mA cm$^{-2}$).

Figure 2a,b shows an optical scanning image of the 6 in. wafers textured with the reference additive (a) and additive 2 (b). These images show inhomogeneous texture marks at the wafer edges in case of the reference sample, where the wafer was in contact with the wet-process carrier. With additive 2, these edge marks are not visible anymore indicating a better wettability during the texturing process and leading to a more uniform texture. Furthermore, photoluminescence (PL) images of the same samples, using the reference additive (Figure 2c) and additive 2 (Figure 2d) and improved contact layers (ITO and metal grid), show an excellent uniformity in the passivation across the whole wafer surface. The inset shows the electrical parameters of this solar cell. Note that the higher $V_{oc}$ compared with the samples in Figure 1b is due to a different wafer material (2 Ω cm$^{-1}$ lower resistivity).

### 3.2. Silicon Surface Cleaning

Proper surface conditioning of the wafer surfaces is of utmost importance to obtain high passivation levels. Possible contamination of the silicon surface with organic or metallic particles can easily induce recombination losses at the heterointerface. Wet chemical conditioning of wafer surfaces influences the electronic interface properties, as reported elsewhere. For that reason, different cleaning processes are analyzed in this section. A process developed by the RCA for microelectronics has become the standard for cleaning silicon wafers, which consists of a two oxidation steps followed by rinsings and HF dips to sequentially remove the organic and metallic impurities of the silicon surfaces. The main drawback of this process is the high consumption of chemicals and the long processing time (≈1 h). Thus, alternative processes which are more environmentally friendly with lower costs than the RCA process have been investigated in the past years, such as wet ozone (O$_3$)-based processes. The ozone-based cleaning processes have a great potential due to the high oxidation rates which etch the organic and metallic residues that the silicon wafer surface might have after the sawing and the texturing steps. In this article, a temperature variation in the ozonized deionized (DI) water-based procedures (room temperature [RT], 30 and 45 °C) is compared with the RCA reference process (done at 80 °C). All ozonized DI water baths for the temperature variation were equally prepared, adding 20 mL HCl and 20 ppm O$_3$. When increasing the bath temperature, to be able to fix the O$_3$ value to 20 ppm, the set value had to be increased slightly for each temperature of the bath, indicating a higher decay of the ozone with temperature. Figure 3a shows effective minority carrier lifetime measurements of four samples equally PECVD coated with the SHJ precursor layers, deposited...
as explained in the Experimental Section, on the four different wet chemically cleaned wafers. The lifetime curves indicate that the ozonized DI water baths at RT and 30 °C provide equivalent passivation as the RCA cleaning. However, when increasing the O$_3$ bath temperature to 45 °C, the lifetime decreased 2 ms absolute compared with the reference RCA sample. As observed by other groups, the ozone decay rate becomes higher when increasing the temperature of the DI-ozonated bath, which agrees with the degrading cleaning properties of the 45 °C bath. Figure 3b shows the median electrical parameters, ($V_{oc}$, FF, and efficiency) of the 14 small-area SHJ solar cells processed in each wafer with the same final clean variation, normalized to the RCA reference. These results confirm that RCA and O$_3$ cleaning procedures both lead to the same passivation, yielding excellent electrical parameters of the finished solar cells. Even though increasing the temperature of the O$_3$ bath is slightly detrimental for passivation (lower lifetime and $V_{oc}$), FF increases slightly, compensating the loss in the final cell efficiency. Note that the lifetime measurement is only in the central area of the wafer while the solar cell parameters are measured on cells distributed across the wafer, which might also add some differences.

3.3. Silicon Surface Passivation by i-a-Si

To know the origin of the limit in passivation due to the PECVD deposited layers, lifetime measurements in symmetric structures were done on dedicated samples. These experiments were done independently of the cleaning experiments mentioned in the previous section; for that reason, all the wafers from this section were textured with the reference additive and cleaned with the standard RCA procedure. Figure 4 shows the lifetime curve of the i/n (black) and i/p (red) symmetrically passivated test wafers. The inset table shows the corresponding median values of lifetime, implied $V_{oc}$, and implied FF of eight identical wafer quarters, showing that the symmetric i/p sample exhibited ≈1 ms and 15 mV lower lifetime and i-$V_{oc}$ values, respectively, than the i/n symmetric sample. To improve the passivation, we replaced the single i-layer by a new a-Si:H passivation stack that consists of a stack of intrinsic layers deposited with different process parameters, i.e., pressure and hydrogen dilution ($R = H_2/SiH_4$).

The new i/p passivating structure (type B) consists of a multilayer stack with three thin i-layers. Lee et al. already used a dual-layered a-Si:H structures for p-type SHJ solar cells, improving the efficiencies due to the better passivation properties of this stack. In our case study, n-type wafers were used and a short hydrogen plasma treatment (HPT) is applied after the deposition of each i-layer in the stack to increase the hydrogen content. Figure 5a schematically compares the new passivating structure with the old one (type A) used as a reference, which is an intrinsic monolayer followed by a long HPT. As already known from the literature, the HPT processes enable improved passivation due to a better hydrogenation of the interface. SHJ solar cell precursors were done using both the new multilayer and the old monolayer structures. In addition, two kind of i-layer materials grown by PECVD were compared, one with a low hydrogen dilution ($R = 1$) and another one with a high dilution ($R = 23$). Table 1 shows the experimental process details of the four samples, using both stacks and materials, and the corresponding lifetime curves are shown in Figure 5b. In blue
are the samples done with the low H₂-diluted i-layers, sample 1 using type A (filled squares) and sample 2 using type B (open diamonds) passivating structures. Comparing those two curves, we can clearly see an improvement in passivation, which correlates with the sample that has an HPT after each i-layer from the stack. The green curves correspond to the type B multilayer structure using the i-layer material with R = 23 in all three layers, sample 3 (open diamonds), and only in the i₂ + i₃ layers, sample 4 (closed circles), this last sample has an i-layer with R = 1 as first layer in contact to the wafer surface (i₁). The lifetime curves show clearly that when using the highly H₂-diluted i-layer in direct contact with the wafer surface, the passivation is detrimentally affected; lifetime decreases by more than 6 ms at an injection level of 10¹⁵ cm⁻³, compared with the sample with the low dilution layer at the interface. This indicates that the layer with R = 23 might induce localized epitaxial growth between the c-Si/a-Si interface, as already observed by several groups with PECVD intrinsic hydrogenated layers deposited with R > 5.[22,23] As reported in the literature, increasing the hydrogen dilution of the a-Si plays a crucial role in passivating the dangling bonds, but an excessive H₂ dilution promotes crystalline (epitaxial) growth of the c-Si wafer surface.[17,24] Thus, using the high H₂ dilution material for layers i₂ and i₃ on top of an i₁ with low H₂ dilution provides the best passivation of all investigated combinations correlating to the higher H₂ content in the bulk of the passivating structure avoiding the epitaxial growth on the c-Si wafer surface thanks to the first i-layer, i₁, which is deep in the amorphous regime, low dilution, and high deposition rate. As mentioned before, after each i-layer from the stack, a short HPT is done to treat the underlying layers, leading to a lower defect density layer that improved the resulting passivation of the silicon wafer. In addition to the passivation improvement in sample 4, electrical measurements of the SHJ solar cells (Figure 6) demonstrate that FF also improves when using the low H₂ dilution material as i₁ combined with the high H₂ dilution material in i₂ and i₃. Figure 6 also shows the implied-Vocc and implied-FF values calculated from the lifetime measurements from the same cell precursors. Both values indicate the potential of sample 4, with >10 mV and almost 2% of i-Vocc and i-FF, respectively, higher than the reference sample 1 using type A passivating structure.

In addition, a thickness variation of the i-layer used in Sample 4 was performed to find the optimum passivating layer. Figure 7 shows the FF versus Vocc parameters from 14 small-area (4 cm², designated area) SHJ solar cells deposited on the same wafer, as shown in the inset and described in the Experimental Section. Figure 7a compares the reference sample (type A i-layer) with a thickness variation of layers i₂ + i₃ (type B i-layer), from 2.2 to 3.7 nm thickness of the total multilayer stack, exhibiting an increase in both FF and Vocc when increasing i₂ + i₃ thickness. The thickness of i₁ is varied from 3.0 to 3.7 nm of total thickness, as shown in Figure 7b. In that case, the JV parameters are not
affected by the thickness variation of this layer, indicating that, once there is a layer that assures an abrupt a-Si/c-Si interface ($i_1$), the layer material that dominates the passivation is the highly $H_2$-diluted one, on the $i_2 + i_3$ layers.

### 3.4. Passivation Improvements in Full-Area SHJ Solar Cells

Implementing the new wet chemistry processes and the new i-layer passivating structure in the full-area, bifacial, busbar-less SHJ solar cells, allowed us to homogeneously scale up the process from small-area ($4 \text{ cm}^2$) to large-area ($244.3 \text{ cm}^2$) cells while keeping losses at a minimum. Table 2 shows the electrical parameters for both size SHJ solar cells, median and best values for the $4 \text{ cm}^2$ solar cells, and the best full-area solar cell (M2). The 6 in. solar cell was measured in a total area ($244.3 \text{ cm}^2$), without using a shadow mask, and in an aperture area ($215.3 \text{ cm}^2$) configuration, using a shadow mask during the measurement that covers 5 mm of the cell edge. This comparison demonstrates that the passivation is excellent in both SHJ solar cells sizes, with $V_{oc}$ values as high as 740 mV for both the median value in small-area and the best total area measurement in full-area solar cells. It is also remarkable that the efficiency in the full-area solar cell measured with an aperture area of 215.3 cm$^2$ reached the same values as the best small-area device, 23.2%, indicating that the improved textured homogeneity and the new i-a-Si stack transfer the passivation gain to the SHJ solar cell efficiency. To avoid shunting at the edges, the rear TCO of the full-area SHJ solar cells is deposited through a metallic mask covering 2 mm along the wafer. 

**Table 2.** IV parameters of small- and full-area (M2) busbar-less and bifacial SHJ solar cells, using the same fabrication processes. The small-area devices are measured with a designated area of $4 \text{ cm}^2$ while the full-area device is measured both with a total area of $244.3 \text{ cm}^2$ and an aperture area of $215.3 \text{ cm}^2$ using a shadow mask covering 5 mm around the wafer edge.

| Cell area | Values | $V_{oc}$ [V] | $J_{sc}$ [mA cm$^2$] | FF [%] | pFF [%] | Eff [%] |
|-----------|--------|--------------|----------------------|--------|---------|---------|
| $4 \text{ cm}^2$ (da) | Median | 0.740 | 38.8 | 79.7 | 84.3 | 22.9 |
| | Best | 0.741 | 38.9 | 80.6 | 84.8 | 23.2 |
| $244.3 \text{ cm}^2$ (t) | | 0.740 | 37.6 | 79.6 | 84.7 | 22.1 |
| $215.3 \text{ cm}^2$ (ap) | Best | 0.738 | 38.5 | 81.4 | 85.4 | 23.2 |
edges. Therefore, the aperture area values show a current density and FF gain of 0.9 mA cm⁻² and a 1.8% rel. compared with the total-area measurements. This exclusion area still needs to be improved to maximize the total-area cell efficiency. This will be done in a following contribution.

4. Conclusions

This article summarizes improvements in rear emitter SHJ solar cells related to wet chemistry and i-a-Si layer depositions. In the wet chemistry experiments, we demonstrated that the additive used in the wet texturing process affects the homogeneity of the texturing and the passivation of the wafer surface. An ozone-based bath has been successfully presented as an alternative cleaning process, which yields lifetime test samples as well as cells with equivalent passivating properties as the standard RCA cleaning, but using a more environmentally friendly method. Regarding the i-a-Si layer, we implemented two different structures to improve the wafer passivation of the i/p side, a monolayer followed by a long HPT and a stack of three very thin layers with a short HPT after each layer. The best passivation structure was the multilayer when using an a-Si:H film grown at low H₂ dilution as first layer in contact with the silicon wafer surface (i₁) and a-Si:H grown with high H₂ dilution as second and third layers from the stack (i₂ + i₃). Finally, implementing all these improvements in passivation together with an enhanced TCO and metal grid, SHJ solar cells with Voc of 740 mV were obtained in both small (4 cm²) and full-area (244.3 cm²) devices demonstrating an excellent scalability of all processes.

Acknowledgements

The authors would like to thank all colleagues at HZB that contributed to this work, especially to Kerstin Jacob, Carola Klimm, Manuel Hartig, Katja Mayer-Stilirich, Holger Rhein, and Matthias Zelt for their valuable help in processing and characterizing. Special thanks to Thomas Gitte from Singulus and Martin Claußnitzer from ICB for their help in wet texturing. In addition, the authors would like to thank GP Solar and ICB for providing the additives for the alkaline texturing solution. They gratefully acknowledge the financial support by the German Federal Ministry of Economic Affairs and Energy within the projects Street (reference #0334275D) and ProSelect (#0324189C). L.K. acknowledges funding from the European Union’s Horizon 2020 research and innovation program under grant agreement no. 727523 (NextBase).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

ozone cleaning, silicon heterojunction solar cells, surface passivation, texturing

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