Solar Energy Materials and Solar Cells 171 (2017) 180–186

Contents lists available at ScienceDirect

Solar Energy Materials and Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Optimized multicrystalline silicon for solar cells enabling conversion efficiencies of 22%

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ARTICLE INFO

Keywords:
Multicrystalline silicon
N-type
Efficiency limiting bulk recombination analysis
TOPCon
Resistivity

ABSTRACT

Multicrystalline (mc) *n*-type silicon has proven to be a suitable substrate for the fabrication of highly efficient mc-Si solar cells. In this paper, we elaborate the impact of base material parameters on the efficiency potential of *n*-type mc-Si solar cells featuring a boron-diffused front side emitter and a full-area passivating rear contact (TOPCon). The electrical material quality can be significantly improved by replacing the standard crystallization process with a seed-assisted growth for crystallization of high-performance (HP) mc silicon. Using high-purity quartz crucibles or larger crucibles in combination with an optimization of the grain boundary area fraction with an adapted seed structure leads to further improvements of the material quality in terms of charge carrier lifetimes. However, not only the charge carrier lifetime, but also the base resistivity is of crucial importance for the efficiency potential depending on the cell concept. Based on experimental data and simulations, we assess the optimal range for the base resistivity and the wafer thickness for *n*-type mc-Si TOPCon solar cells. With the optimal material parameters, an “efficiency limiting bulk recombination analysis” (ELBA) reveals an efficiency potential in the range of 22.5% for *n*-type mc-Si TOPCon solar cells. Finally, we fabricated TOPCon solar cells based on optimized *n*-type HP mc-Si substrate and demonstrate a certified efficiency of 21.9%, which is the highest efficiency reported for multicrystalline silicon solar cells so far.

1. Introduction

With progress in crystallization techniques, such as seed-assisted growth for the fabrication of so called “high-performance” (HP) multicrystalline (mc) silicon [1], the material quality of mc-Si wafers has increased significantly in the past years, mainly due to a reduced density of recombination-active dislocation clusters. Higher quality mc-Si substrates in combination with advancements in solar cell architectures resulted in a new certified world record mc-Si solar cell featuring an efficiency of 21.3% [2,3]. Like all industrially fabricated mc-Si solar cells, this record cell is based on *p*-type mc-Si substrate. However, the highest silicon solar cell efficiencies on monocrystalline silicon are achieved on *n*-type substrate, such as the current records of 26.6% [4] and 25.3% [5] for heterojunction and homojunction cells, respectively. Due to the smaller impact of many metal impurities, such as interstitial iron, on the electrical material quality [6] and the absence of the boron-oxygen-related degradation [7,8], *n*-type doping could also be a promising option for multicrystalline silicon solar cells, if an appropriate cell concept is provided. We have recently shown that the TOPCon cell concept [9] featuring a boron-diffused front side emitter and a full-area passivating rear contact is applicable to *n*-type mc-Si substrate and demonstrated an efficiency of 19.6% [10]. A main limitation of this solar cell was the poor optics of the isotextured front surface, which capped the cell limit without bulk recombination losses to approximately 21%. With the development of a plasma-etched “black-silicon” texture in combination with further technological advancements, a cell process with a concept-related efficiency limit above 23% (without defect recombination losses) has been developed, which is applicable for the fabrication of *n*-type mc-Si TOPCon solar cells [11]. With regard to this high cell limit, the reduction of material-related efficiency losses in the mc-Si substrate becomes of utmost importance, which is the focus of this work. Based on experimental results in combination with simulations, we quantify efficiency gains from improvements of the crystal structure and the optimal choice of

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http://dx.doi.org/10.1016/j.solmat.2017.06.005
Received 12 April 2017; Received in revised form 2 June 2017; Accepted 5 June 2017

Table 1
n-type mc-Si wafers.

	Crystallization	Crucible	Coating	Resistivity (Ω cm)
A	standard	G2 standard	ISE	2.0
B	HP	G1 HPC	Vesuvius	1.0
C	HP	G2 standard	Vesuvius	10.8
D	HP optimized	G2 standard	Vesuvius	0.75

parameters for resistivity and wafer thickness. Thus, we present a concept how to optimally exploit a cell's efficiency potential by minimizing material-related efficiency losses.

2. Material and methods

The development of *n*-type mc-Si materials at Fraunhofer ISE and their evaluation for the use in high-efficiency solar cells comprised standard directional solidification processes with nucleation of initial grains directly at the crucible bottom (“standard process”) as well as materials with nucleation at seed materials (“high-performance process”). The ingots to be evaluated had either the laboratory size G1 equivalent to 15 kg of Si feedstock (supplied by Wacker Polysilicon) or laboratory size G2 equivalent to 75 kg. From both types of ingot center bricks were cut and further processed into wafers. In this study, we included four different *n*-type mc-Si materials, one from a standard crystallization process and three from high-performance (HP) crystallization processes with variations in the seed structure, the crucible systems as well as the base resistivity (*cf.* Table 1). Wafers were taken from the upper third of each ingot: wafer “A” stems from a standard crystallization process in a G2 size crucible of standard purity coated at ISE, wafers “B-D” are HP mc-Si wafers originating from seed-assisted growth crystallization processes. Material “B” was crystallized in a G1 size crucible of high purity (Vesuvius “high purity crucible” HPC with Vesuvius coating), wafers “C” and “D” in G2 size crucibles (Vesuvius standard crucibles with Vesuvius coating). Wafers “C” and “D” differ strongly in their resistivity. Further, for material “D” the seed-assisted growth was adapted in order to obtain an optimized grain boundary area fraction, featuring a sufficiently high amount of grain boundaries for the suppression of dislocation clusters, but favouring the growth of large grains at the same time. This was done by using a plate cut from a high-performance mc-Si ingot as seed layer in the crucible bottom instead of granular silicon. This plate features an optimal distribution of grain sizes such that largest possible grains develop along the entire ingot height without the emergence of recombination active dislocation clusters. For the high-performance wafers investigated here, the median grain sizes increase from ~ 9 mm^2 (“B”) to ~ 24 mm^2 (“C”) and ~ 36 mm^2 (“D”).

It should be noted that the crystallization processes in terms of crystallization times and cooling ramps are identical for materials “C” and “D”, whereas the other two differ either due to a different crucible size (G1 for material “B”) or due to a different crystallization technique (“standard” instead of high-performance for material “A”). Of course, this also comes along with variations in the impurity content of the different materials [12]. As a change in the crystallization technique always requires changes in the processing parameters, it inherently involves differences in the impurity content of the crystallized materials. Thus, the results presented in this paper comprise the impact of combined effects arising from changes in the crystallization techniques.

The wafers were processed to lifetime samples by applying the high-temperature steps of a TOPCon solar cell process sequence, i.e. a boron diffusion at 890 °C and an annealing step at 800 °C (both for 1 h) (*cf.* Fig. 1). This ensures that the material quality of the lifetime samples corresponds to the material quality of the final cell and allows for an analysis of the material related efficiency losses. To obtain an injection-independent surface passivation, the wafers were passivated with SiN$_x$-films. Injection-dependent photoluminescence (PL) imaging calibrated by modulated PL [13] allows for spatially resolved characterization of the bulk minority charge carrier lifetime τ_{bulk} (in the following referred to as “bulk lifetime”) and, in combination with PC1D [14,15] cell simulations, for a prediction of the cell efficiency potential by an “Efficiency limiting bulk recombination analysis” (ELBA) [16]. The latter additionally enables a detailed loss analyses as presented in [17]. The parameters for the PC1D model were taken from recombination current prefactor (J_0) measurements of the TOPCon rear side ($J_{0,\text{rear}}$ = 7 fA/cm^2), the black-silicon boron-diffused front side ($J_{0\text{e}}$ = 65 fA/cm^2), reflection measurements of the front side (weighted reflection = 1%) [11], and base resistivity measurements of the mc-Si material. Furthermore, a series resistance of 0.55 Ω cm^2 was assumed. This leads to a cell limit without bulk recombination losses of 23.1%.

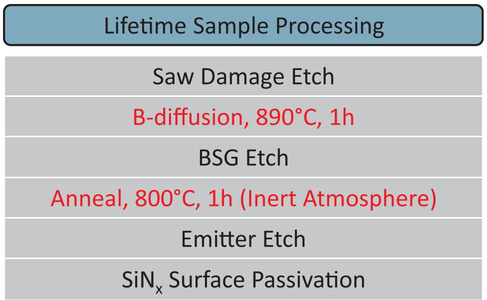

Fig. 1. Lifetime sample processing sequence. By choosing the high-temperature steps of a TOPCon solar cell process, we obtain lifetime samples with material quality corresponding to that of the TOPCon solar cells.

3. Results

3.1. Bulk lifetime

For a first evaluation of the material quality, Fig. 2(a) shows PL images of τ_{bulk} obtained at an irradiation of ~ 0.05 suns (~ corresponding to injection conditions at maximum power point, MPP) together with the average lifetime across the image. In the following, all average lifetime values are square root harmonic means, indicated by $\tau_{1/L}$. Wafer “A” from the standard crystallization process features some large grains of high bulk lifetime close to 1 ms. The clear drawback of this material are large areas featuring a high density of strongly recombination-active structural crystal defects such as grain boundaries and dislocation clusters, which limit the area averaged lifetime to 138 µs. The strongly recombination-active dislocation clusters can be avoided by seed-assisted growth, delivering high-performance multicrystalline silicon. In combination with use of a high purity crucible, this leads to smaller grains with higher bulk lifetimes. Thus, the average bulk lifetime is increased to 403 µs (wafer “B”). While the best grains of this material feature bulk lifetimes close to 2 ms, the average is still limited by a rather large amount of recombination-active grain boundaries. To reduce their impact, we followed two approaches: Material “C” features a higher resistivity (*cf.* Table 1), which leads to a reduced recombination at metallic precipitates [18], and for material “D” we adapted the seed-assisted growth such that an optimized grain boundary area fraction is obtained: on the one hand, it delivers a sufficiently high amount of grain boundaries for the suppression of dislocation clusters, on the other hand it favours the growth of still large grains.

Both materials “C” and “D” feature high inner grain bulk lifetimes exceeding 2.5 ms. Despite the virtually identical impurity content of these two materials and the even better crystal structure of wafer “D”, the average bulk lifetime of wafer “C” (913 µs) is significantly higher than that of wafer “D” (634 µs), which is attributed to reduced recombination in the high resistivity wafer “C” due to a combination of the following effects:

(1) Lower intrinsic recombination (radiative and Auger).
(2) Lower Shockley-Read-Hall (SRH) recombination: A lower base