Growth and characterization of multicrystalline silicon ingots by directional solidification for solar cell applications

R. Bairava Ganesh*, Birgit Ryningenb, Martin Syvertsenb, Eivind Øvrelidb, Ivan Sahaa, Harsharn Tathgarc and G. Rajeswarana

aMoserbaer Solar Limited, 66-B, Udyog Vihar, Greater Noida, G.B. Nagar, U.P-201 306, India
bSINTEF Materials and Chemistry, Department of Metallurgy, Alfred getz v 2, Trondheim, Norway
cUmoe AS, Fornebuveien 84, N-1366 Lysaker, Norway

Abstract

This study is focussed on the growth of multicrystalline silicon ingots with large grains by controlling the silicon melt cooling rate to initiate dendritic nucleation in the initial stage of the solidification. Two ingots were grown with different undercooling rates and compared with a reference ingot grown by standard cooling conditions. All ingots were grown in a lab scale directional solidification system. The wafers cut from all three ingots have been characterized for resistivity, minority carrier lifetime and dislocation density measurements by four point probe, quasi steady state photo conductance and PV Scan, respectively. The wafers were converted into solar cells and their electrical parameters have been measured. The cells fabricated from ingot 2 show slightly higher efficiencies in comparison with ingot 1 and the reference one. The present cooling rate was not enough to initiate the dendrite nucleation in the beginning of the solidification. Hence, there is no significant difference was observed in the crystal quality of the grown ingots 1 and 2.

* Corresponding author. Tel.: +91-120-4658200; Fax.: +91-120-4658164
E-mail address: bairava.ganesh@moserbaer.in

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1. Introduction

In photovoltaic industry around 90% of all solar cells produced are based on crystalline silicon wafers and in recent past multicrystalline silicon (mc-Si) is considered to be the most promising type of material capable to achieve the low manufacturing cost of the solar cell fabrication [1]. Directional solidification is commonly used technique for producing multi-crystalline silicon ingots because of its highly columnar grain growth nature with planar interface [2,3]. It is well known fact that lower efficiency in mc-Si solar cells is mainly due to the presence of crystal defects such as dislocation, grain boundaries and impurity precipitations. Recombination of the minority carriers at the dislocations and grain boundaries which are decorated with impurities reduces the conversion efficiency of the mc-Si solar cells drastically [4]. The inferior performance of the mc-Si solar cells compared to monocrystalline silicon is also due to the presence of many smaller crystallographic grains and their grain boundaries [5]. Recent studies show that dendritic nucleation at initial stages of growth process leads to larger grain size along with more homogeneity of grain orientation than randomly nucleated ingot [6]. Fujiwara et al. have studied controlled nucleation of directionally solidified mc-Si ingots on a laboratory scale, and reported that dendritic growth in the nucleation phase will occur under specific conditions. The solar cells fabricated from those ingots were reported to have high conversion efficiencies as comparable to monocrystalline silicon [7].

In the present investigation, attempts have been made to initiate dendritic nucleation at start of growth process by controlling the undercooling and results are presented. The multicrystalline structures and solar cell properties of these ingots were compared and the results are given in detail.

2. Experimental

Three ingots were grown in this project; one with standard casting parameters (labelled Ref Ingot) and other two ingots grown with higher cooling rate to get higher undercooling before solidification (labelled Ingot 1 and Ingot 2). Besides the cooling rate change, we ensured that all other parameters, which have significant contribution on quality of ingot and process, were fixed. Important parameters are listed below.

- Dopant concentration to achieve target resistivity of 0.75 $\Omega \text{cm}$ in the middle of the block
- Electronic grade crucibles from same shipment
- Same UBE $\text{Si}_3\text{N}_4$ coating powder for all growth runs
- Same person for crucible coating and charge loading
- Growth was carried out by without rotation and lowering of crucible in all runs
- Growth runs in same hot zone

To achieve undercooling for Ingot 1 and Ingot 2, the temperature was maintained for one hour holding time after melt down for better melt homogenisation, and then it was decreased by 100°C per hour instead of 50°C per hour which was used for standard cooling rate of ref ingot. The ingots were cut into blocks, further sliced into wafers and side cuts were grinded. Resistivity and minority carrier life time was measured by Four point probe and Quasi Steady State Photo Conductance (QSSPC) [8] respectively. Two neighbouring wafers were taken out of every 10th wafer throughout the whole height of all three ingots. The two wafers from the bottom of the ingots were polished and one of them was etched by Sopori etch for dislocation mapping by PV Scan [9,10]. The wafers from the two ingots along with
wafers from the reference ingot were converted into cells at Moserbaer Solar Limited (MBSL) India. The solar cell fabrication process flow is shown in figure 1.

![Solar cell fabrication process flow](image)

**Figure 1.** Multicrystalline silicon solar cell fabrication process flow

### 3. Results

Figure 2a shows the resistivity variation of all three ingots along growth direction. Ref ingot, grown with standard settings, showed higher resistivity at top than further down, while ingots 1 and 2, grown with higher under cooling rate, had the variation in the resistivity. Figure 2b indicates the lifetime variation in all three ingots, where Ref ingot measured at two places and Ingot 1 are showing nearly same value and Ingot 2 is showing low values.

![Graphs](image)

**Figure 2.** (a) Resistivity as a function of height; (b) Minority carrier lifetime of grown ingots
Figure 3a shows the comparison of the dislocation densities of the three ingots measured by PV Scan. From the PV Scan data there seems to be no significant difference between the three ingots. Optical scans of polished and etched bottom wafers of Ref ingot and Ingot 2 were also investigated and shown in figure 3b and 3c, respectively. Though there was no clear difference between the major portions of the ingots, however, a little difference is observed at the very top of ingots, where ingot 1 has a high number of small grains than other ingots.

3.1 Solar cell performance

After converting these wafers into cells, the electrical parameters of these cells were compared with the cells which are fabricated from the standard wafers used in the manufacturing. The relative variation in the electrical parameters of the fabricated cells is shown in figures 4a, 4b, 4c and 4d. All fabricated cells have shown inferior performance in comparison with the cells from the standard wafers. In this study the ingots measure 25 cm in diameter, 10 cm height and the charge is only 12 kg. Due to the high surface to volume ratio, surface effects are more pronounced than in industrial systems. This results in lower minority carrier lifetime both in bulk and near the surfaces respectively. It is presumed that the incorporation of metallic impurities from crucible and coating materials is more pronounced in ingots grown by a lab scale system, than the ingots grown in industrial scale furnaces, where majority portion of ingots are relatively with low impurity concentration. Hence these ingots shown inferior performance in comparison with the standard wafers used in manufacturing. The cells fabricated from wafers of ingot 2 had shown high conversion efficiencies in comparison with wafers from the other ingots.
4. Discussion

Wafers from three ingots with different cooling parameters were investigated with respect to dislocation density, microstructure, minority carrier lifetime and solar cell performance. The lifetime measurement showed that ingot 2 had a lower minority carrier lifetime than other ingots which may be due to unintentional weak spot in the coating which resulted incorporation of metal impurities from the crucible during solidification. During solar cell processing, the phosphorus diffusion induced gettering of metal impurities will improve the electrical parameters of the solar cell. Ingot 2 had achieved proper gettering during solar cell processing since it effectively removes the remnant contaminations such as Fe, Cr and Cu from the wafer bulk. Gettering helped in increasing carrier lifetime, thus ingot 2 achieved slightly higher solar cell efficiency than the other ingots. In the case of ingot 1, lower efficiencies were observed in the cells made from the wafers cut at top portion of ingot, where smaller grains are spotted. The smaller grains seen at the uppermost part of an ingot is a frequently occurring phenomenon and is most likely caused by cooling from the gas inlet situated just above the melt surface. This cooling effect along with higher metallic impurity concentrations in some cases makes the final part of solid-liquid interface solidify faster than rest of ingot, which normally resulted to smaller grains at top of an ingot. This phenomenon caused a difference in the solar cell electrical properties of ingots 1 and 2, other wise they were solidified in the same conditions. From the optical scan of bottom wafers, it is clear, from the micro structure, that the desired dendrite nucleation was not achieved in ingot 1 and 2. In addition earlier report implies that an undercooling of approximately $\Delta T=10^\circ$ K to be required to induce dendrite growth in the initial stage of solidification which was not achieved [11]. A cooling rate of 100°C per hour is thereby not sufficient to achieve the undercooling necessary to initiate dendrite nucleation in the pilot scale Crystalox furnace.

5. Conclusion

We investigated the effect of silicon melt cooling rate in directional solidification process for multicrystalline silicon ingot. Two ingots with fast cooling rates and one reference ingot using standard cooling condition have been grown in this project. From the lifetime measurements, wafers sliced from ingot 2 grown with higher cooling rate, have lower lifetime in comparison with the other ingots. The PV Scan studies indicated that all the grown ingots have similar dislocation density values. The wafers were converted into solar cells and their electrical parameters were measured. The solar cell data show that the wafers from ingot 2 have slightly higher efficiency due to gettering process during cell fabrication. Smaller grains were observed in the uppermost part of the ingot 1 hence lower conversion efficiencies
were obtained in these solar cells. In this study, significant differences between the ingots were not observed due to the change in cooling rate. A cooling rate of 100°C per hour was not enough to initiate the desired dendrite nucleation in the beginning of the solidification to get bigger grains in the multicrystalline silicon ingots. Future experiments are in progress to investigate the effect of undercooling in detail.

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References