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Evolution of grain structure and recombination active dislocations in extraordinary tall conventional and high performance multi-crystalline silicon ingots

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ABSTRACT

In this work one high performance multi-crystalline silicon ingot and one conventional multi-crystalline silicon ingot, each with an extraordinary ingot height of 710 mm, were replicated by the successive growth of eight G1 ingots to evaluate the potential advantage of extraordinary tall HPM ingots in industrial production. By analyzing different grain structure parameters like mean grain size, grain orientation and grain boundary type distribution as well as the recombination active dislocation area over the complete ingot height, it was observed that the material properties strongly differ in the initial state of growth for the two material types. However, at ingot heights above 350 mm, the difference has vanished and the grain structure properties for both materials appear similar. It is shown that the evolution of the grain structure in both material types can be explained by the same grain selection and grain boundary generation/annihilation mechanisms whereas the current grain structure determines which mechanisms are the most dominant at a specific ingot height. Since the grain structure directly influences the dislocation content in the silicon material, also the recombination active dislocation area becomes equal in high performance and conventional multi-crystalline silicon material at ingot heights above 350 mm. From these results it is concluded that the advantage of high performance silicon material is limited to the first grown 350 mm of the ingot.

1. Introduction

Nowadays, directionally solidified multi-crystalline (mc) silicon is one of the most used materials for the production of silicon solar cells. For a few years, the conventional mc-silicon with the silicon melt solidifying directly at the Si_3N_4 releasing coating at the crucible bottom, is being replaced more and more by the so called "high performance mc-silicon" or HPM-silicon which was firstly reported by Sino-American Silicon Productions Inc. (SAS) in 2011 [1,2]. The HPM material, which is produced either by nucleation on a feedstock particle layer [2–6] or more recently on a structured crucible bottom/functional coating [7–9], is characterized by a very fine initial grain structure (mean grain size $< 4\ mm^2$), a homogeneous orientation distribution (small coefficient of variation $CV_{GO} < 1.5$, explanation see Section 2) and a high length fraction of random grain boundaries > 60% (see e.g. [10]). These grain structure properties lead to a significantly smaller amount of recombination active dislocations and therefore result in ~0.5% absolute higher solar cell efficiencies [2,10] in comparison to the conventional mc-silicon which is characterized by larger grains containing many dendrites and twins.

Recently, it was observed by Lehmann et al. [10], who investigated the grain structure properties of several industrially grown HPM and conventional mc-Si ingots, that the difference in grain structure properties and also the amount of recombination active defects between the two material types was not as significant at the top region as it initially occurs at the bottom region of the ingots. From this observation, it seems that the advantage of the HPM silicon mainly occurs in the lower parts of the ingot where the difference of the HPM grain structure to the conventional mc grain structure is largest.

Currently, silicon ingots produced in industrial scale exhibit an ingot height of about 250–400 mm leading to the above described benefit of HPM silicon. In order to clarify if there is some advantage of the growth of industrial HPM ingots with an ingot height > 400 mm within the present work, the evolution of grain structure properties and the recombination active dislocation area in extraordinary tall silicon ingots was investigated. For that purpose one HPM silicon ingot and

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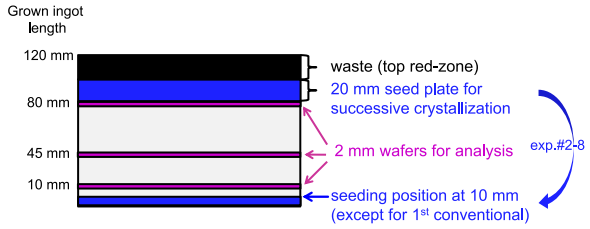

Fig. 1. Sketch of the procedure of growing extraordinary high conventional and HPM silicon ingots with a total ingot height of 710 mm by successively growing of eight G1 ingots. In the case of the 1st grown conventional G1 silicon ingot, there is no initial seeding layer.

one conventional mc-silicon ingot, each with an extraordinary ingot height of 710 mm, were replicated by the successive growth of eight G1 ingots.

2. Experimental setup and characterization

A crystallization furnace, which allows to grow G1 silicon ingots with dimensions of 220×220×130 mm^3 and a weight of 15 kg, was used for the experiments [11]. For all experiments, standard fused silica crucibles with a standard Si_3N_4 release coating and polysilicon feedstock from Wacker were applied. To obtain silicon ingots with a "total" ingot height of 710 mm, successive growth of eight G1 silicon ingots was carried out. The relating procedure is sketched in Fig. 1. For the first HPM as well as for the first conventional G1 ingot the crucible was filled completely with the silicon feedstock. In the case of the HPM ingot a 20 mm seeding layer containing only small silicon chips was placed on the crucible bottom. During the crystallization process 10 mm of this seeding layer was kept unmelted in order to achieve a very fine HPM grain structure by solidifying the silicon melt directly on the feedstock particles. In the case of the conventional ingot, the feedstock was completely melted resulting in a more coarse and dendritic grain structure by solidifying on the standard Si_3N_4 coating at the crucible bottom. Besides the small difference during the melting phase, the crystallization processes for both the HPM and the conventional silicon ingot were quite similar with a growth rate of about 1 cm/h and an almost flat solid-liquid interface shape during the whole growth process.

After growing the first ingots, 20 mm thick slices were cut from 80 mm to 100 mm grown ingot length and used as seed plates for the second crystallization experiments. This step was repeated six times until eight silicon ingots were grown corresponding to a "total" growth length of 710 mm designated as "total ingot height" in the following text.

In terms of characterization, three horizontal wafers with an area of 156×156 mm^2 and a thickness of 2 mm were prepared from the center of each ingot at 10 mm, 45 mm and 80 mm grown ingot length. After mechanical preparation of the wafer surface, the grains of the wafers were automatically detected in full wafer scale by an optical system (GEMINI tool from Intego [12]) which is based on reflectivity. Twenty images of the wafer surface are taken by illuminating the wafer by LED modules placed at different angles to the wafer surface. Subsequently, the position and the size of each grain are determined by an automatic image processing step. The minimum detectable grain size for the present investigations was around 0.075 mm^2. A description of the grain size distribution was done by using the so-called coefficient of variation $CV_{grain\ size}$ (CV_{GS}) which is defined by the ratio of the standard deviation of the mean grain size and the mean grain size itself. This value, often used in statistics, reflects the homogeneity of the grain sizes means the smaller the CV value, the more homogeneous is the grain size distribution.

The crystallographic orientation for grains larger than 3 mm^2 was determined in a second step by using a Laue scanner [12]. This system irradiates each grain with white x-rays and detects the resulting Laue pattern. Analysis of the Laue pattern is done automatically by the system. As a result, the grain orientation perpendicular to the wafer surface, which nearly equals the growth direction, is obtained. Furthermore, the grain boundary type is determined if the grain orientation of two neighboring grains was measured. The measured grain orientations were simplified to *hkl* values up to 20. In the following, the results are depicted as circles drawn into inverse pole figures (IPF). The center of a circle represents the grain orientation itself and its diameter is proportional to the area fraction of that grain orientation. The grain orientation distribution is also described by using the coefficient of variation $CV_{grain\ orientation}$ (CV_{GO}). In this case, the CV_{GO} is defined as the standard deviation of the area fractions of the grain orientations divided by the mean value.

After measuring the grain orientation, a polishing etch containing $HF:HNO_3$ was applied on the same wafers and subsequently band to band photoluminescence imaging (PLI) was performed to investigate the area fraction of recombination active dislocations. The PLI measurements were done with the OPTECTION imaging tool [13] using a 175 W laser (wavelength λ 790 nm) and a 40 s exposure time. The area fraction of recombination active dislocations was determined by post-processing and analysis of the resulting image.

3. Results

First, the results gained from the grain structure analysis are presented and discussed. Fig. 2 shows the mean grain size as well as the coefficient of variation CV_{GS} over the total ingot height for the 710 mm G1 HPM ingot and the 710 mm G1 conventional ingot, respectively. As expected, the mean grain size at the ingot bottom is significantly larger for the conventional ingot than for the HPM ingot. With increasing ingot height the mean grain size increases in both ingots. Additionally, the difference between the two curves becomes smaller and vanishes within the margin of error at about 150 mm. Over the further ingot height up to 710 mm, the mean grain size remains constant for both ingot types.

This can also be seen from the grain structure images obtained by the GEMINI tool, which are shown in Fig. 3. While there is a large difference in the initial grain structure of both ingot types at 10 mm grown ingot length, the grain structures becomes more and more similar with increasing ingot height and no significant difference can be observed by eye at 710 mm.

In the case of the coefficient of variation CV_{GS} a similar trend is observed, but even more clearly. Again, the CV_{GS} values are distinctly different at the ingot bottom for both material types. While the conventional ingot exhibits a high initial CV_{GS}=6.6 which decreases with increasing ingot height, the HPM ingot has a small initial CV_{GS}=1.5 slightly increasing with increasing ingot height. After about 330 mm ingot height, both level at CV_{GS}≈3 and exhibit an almost constant trend up to the ingot top at 710 mm. Again, this is comprehensible if we look on the grain structure images in Fig. 3. The initial grain size distribution is quite inhomogeneous in the case of the conventional ingot due to several dendrites and twins. Oppositely, the initial HPM structure exclusively consists of very small and nearly isometric grains without any twins or dendrites. With increasing ingot height the grain size distribution for both ingot types looks more and more similar.

For comparison, the correspondent values for an industrially grown HPM ingot with an ingot height of about 300 mm were added to the curves in Fig. 2. The curves for the 300 mm industrial HPM ingot correlate well to the values of the first 300 mm of the successively grown 710 mm ingot. Only in the second ingot half, the mean grain size is slightly higher for the industrial ingot. This good agreement validates that the used method of the successive growing of several 130 mm tall ingots yields highly comparable results with respect to growth within one process as it is done in industry. This allows us the transfer of the results from our experiments to extraordinary high industrial ingots.