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Influence of different seed materials on multi-crystalline silicon ingot properties

C. Reimann [a,b,*], M. Trempa [a], T. Lehmann [b], K. Rosshirt [a], J. Stenzenberger [c], J. Friedrich [a,b], K. Hesse [c], E. Dornberger [c]

[a] *Fraunhofer Institute for Integrated Systems and Device Technology (IISB), Schottkystrasse 10, 91058 Erlangen, Germany*
[b] *Fraunhofer THM, Am St.-Niclas-Schacht 13, 09599 Freiberg, Germany*
[c] *Wacker Chemie AG, Johannes-Hess-Straße 24, 84489 Burghausen, Germany*

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ABSTRACT

Different silicon feedstock materials, Single Crystalline Crushed (SCS), Fluidized-Bed-Reactor (FBR) and Siemens (SIE) feedstock, were used as seeding layer for growing cylindrical shaped, high performance multi-crystalline ingots with a weight of 1.2 kg. Within the investigations a systematic variation of the particle size of the seeding material in the range of <1 mm up to 15 mm was performed. Grain size, grain orientation, and grain boundary type were evaluated at different ingot heights. These results show clearly, that the microstructure size, respectively the particle size for the crushed single crystalline material, determines the resulting grain structure in the ingot near the seeding position. If the microstructure size is equal to the particle size, as it is the case for the SCS material, the particle size has a significant influence on grain size, grain orientation, and grain boundary distribution. With increasing average particle size of the SCS seed material the grain size increases, the grain orientation distribution becomes less uniform, and the random grain boundary length fraction decreases. If the microstructure size is smaller than the particle size, as it is the case for FBR and SIE feedstock materials, the particle size has no influence on the initial grain structure of the ingot. For FBR and SIE seeding material, small grains, with a homogeneous orientation distribution and a high random grain boundary length fraction are obtained. Therefore, all FBR and all SIE seeding materials, as well as the SCS with particle size <1 mm, show lowest fractions of defected areas at about the same level which were determined by etch pit analysis.

1. Introduction

Silicon is the dominant absorber material for solar cell application. Multi-crystalline silicon (mc-Si) has an actual market share of about 50% [1]. The state-of-the-art crystal growth technology in industry for producing mc-Si ingots is the directional solidification (DS) process, using silicon feedstock which is placed in a Si_3N_4 coated fused silica crucible. For the so-called high-performance (hp) process a layer of special silicon feedstock material is typically placed on the crucible bottom, which is partially melted during the initial phase of the DS process. The preserved feedstock acts as a seeding layer for the subsequent epitaxial like crystal growth. The resulting multi-crystalline silicon ingots exhibit typically a fine grained structure, combined with a low area fraction which is electrical active. Recent work by Lehmann et al. [2] shows clearly that the fine grained structure in combination with a homogenous orientation distribution and a high fraction of random grain boundaries are the ingredients for solar cell efficiencies of up to 18.8%, and even higher depending also on the solar cell process.

Several research groups and companies [3–10] are working in the field of hp ingot crystallization and characterization. So far the question of the influence of the used feedstock material as seeding layer on the resulting ingot properties was not answered in detail.

This contribution deals with a systematic variation of the silicon seeding material. Single Crystalline Crushed (SCS), Fluidized-Bed-Reactor (FBR), and Siemens (SIE) feedstock were used in a particle size range from <1 mm up to 15 mm as seeding layer in directional solidification experiments in a lab scale furnace. The influence of the different seeding materials on the resulting grain structure (grain size, grain orientation and grain boundary configuration), and on the etch pit density of the directionally solidified hp ingots was investigated.

* Corresponding author at: Fraunhofer Institute for Integrated Systems and Device Technology (IISB), Schottkystrasse 10, 91058 Erlangen, Germany.
E-mail address: Christian.reimann@iisb.fraunhofer.de (C. Reimann).

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2. Experimental methods

A lab scale Vertical Gradient Freeze (VGF) furnace [11] was used to grow cylindrical ingots with a diameter of 100 mm, a height of about 65 mm and a resulting ingot weight of 1.2 kg (compare Fig. 1 left). Commercially available, standard Si_3N_4 coated fused silica crucibles were used for the investigations. The crystallization process can be described by a nearly flat interface shape with some small concave interface deflection at the crystal edges with a typical solidification rate of about 1 cm/h during the whole crystallization step. The height of the seeding layer in all cases was in the range of 2 cm.

As seeding layer material Single Crystalline Crushed (SCS), Fluidized-Bed-Reactor (FBR), and Siemens (SIE) feedstock were used in a particle size range from <1 mm up to 15 mm. Table 1 gives an overview of the different particle sizes of the various seeding materials.

SIE feedstock was used as standard feedstock load above the seeding layer in all cases.

In experiment series A the particle size of the SCS material was varied, while in experiment series B the particle size of FBR and SIE material was altered.

After crystallization the lab scale ingots were half-cut through the center. Subsequently the grain structure was investigated on a vertical cut, as well as on horizontal cuts 5 mm below, and 5 mm as well as 25 mm above the seeding position in an as-cut state (compare Fig. 1).

The analytic tools used for the grain size, grain orientation, and grain boundary type detection were described elsewhere [12,13]. For evaluating the results the coefficient of variation CV was determined, which is defined as standard deviation of the considered data divided by the mean value of the considered data. The CV value gives an information about the uniformity of grain size ($CV_{\text{Grain Size}=\text{GS}}$), and grain orientation ($CV_{\text{Grain Orientation}=\text{GO}}$). The detection error for the grain size is 10% relative, for the $CV_{\text{Grain Size}=\text{GS}}$ value it is 5% relative, and for the $CV_{\text{Grain Orientation}=\text{GO}}$ value it is 20% relative.

Furthermore, etch pit density (EPD) measurements were performed for the 25 mm cut after polishing and subsequent Secco etching [14] by reflected-light microscopy using subsequently an automatic mapping and afterwards EPD counting routine.

For comparison, results were added for a crystallization procedure without a silicon seeding step, which will be named in the following as conventional multi.

Table 1
Overview of the different particle sizes of the various seeding materials SCS, FBR and SIE and their sample names in the further text.

Seed material	Sample name and particle size				
SCS	SCS1 10–12 mm	SCS2 3–6 mm	SCS3 <3 mm	SCS4 <1 mm	
FBR	FBR1 5–10 mm,	FBR2 0.5–3 mm	FBR3 1–3 mm	FBR4 1–2 mm	FBR5 0.5–1 mm
SIE	SIE1 0.2–15 mm	SIE2 0.2–7 mm			

3. Results and discussion

Fig. 2 shows a schematic picture for the used SCS, FBR, and SIE particles. The particle size in combination with the crystallographic microstructure size is indicated. It can be seen clearly that for the used SCS material the particle size equals with the microstructure size independently of the used particle size, due to the fact that it is a crushed single crystal. For the used FBR and SIE materials the particle size is unequal to the microstructure size, due to the fact that it has a polycrystalline microstructure. The microstructure of the used SIE material consists of several larger grains with 100 μm in diameter and many small grains with a

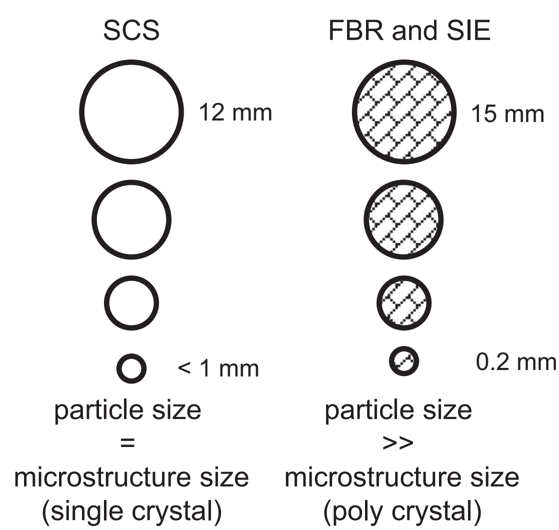

Fig. 2. Schematical drawing for the used particles in combination with the microstructure size.

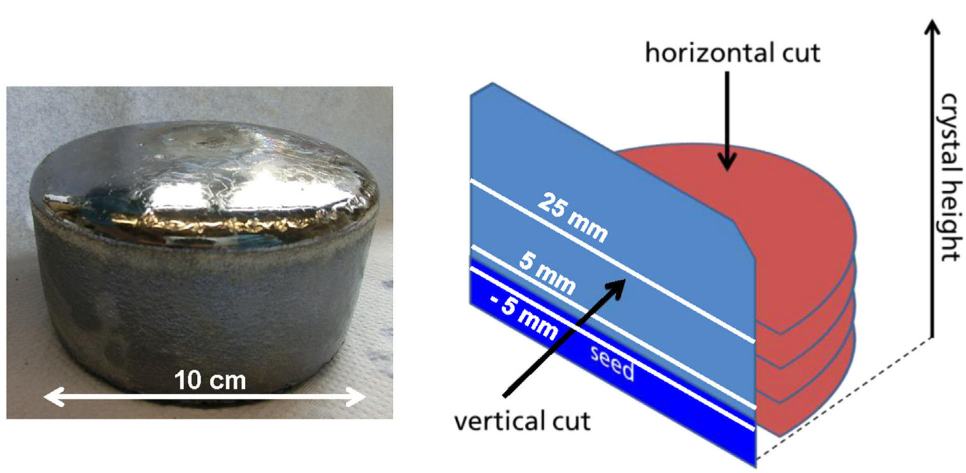

Fig. 1. Multi-crystalline silicon ingot, solidified in the Vertical Gradient Freeze (VGF) furnace (left) and cutting scheme for further sample characterization (right).