



About the formation and avoidance of C and N related precipitates during directional solidification of multi-crystalline silicon from contaminated feedstock

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Abstract

Multi-crystalline silicon ingots with a diameter of 6cm and a height of 4–5cm were directionally solidified in a laboratory scale crystal growth facility within a Si₃N₄-coated fused silica crucible with a growth rate of 1 cm/h for two different conditions of convective transport in the melt. The feedstock quality was varied from “pure” (electronic grade) to highly carbon-contaminated and highly nitrogen-contaminated. It will be demonstrated that under certain convective process conditions even for a highly contaminated feedstock (C and N) a mc silicon ingot can be grown which has an axially and radially homogenous carbon and nitrogen concentration in the range 3–4×10¹⁷ C-atoms/cm³ and 2–4×10¹⁵ N-atoms/cm³, respectively, and is free of SiC- and Si₃N₄-precipitates in the bulk.

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Introduction

Nowadays directional solidification is the dominant technology for producing multi-crystalline (mc) silicon ingots for solar cell applications. Beside process optimization for improved material quality, cost reduction steps by comparable solar cell efficiencies are also getting into the focus. The use of less expensive silicon raw material e.g. upgraded metallurgical grade, as well as an increased silicon throughput per time unit for a crystal growth run are dominating the developments worldwide [1], [2].

The properties of mc silicon like minority carrier lifetime or diffusion length, which are important properties for the solar cell efficiency, are correlated to the content and the occurrence of C, N, O, metals and unintended dopants as well as structural defects (dislocations, grain boundaries) and its interaction [3], [4], [5]. C and N contamination of the silicon melt lead to the formation of SiC- and Si₃N₄-precipitates in the melt and in the crystal, respectively [6], [7], [8], [9], [10], [11], [12], [13]. These precipitates induce wire cracking during the wafer sawing process [14] and lead to a failure of the solar cell due to the occurrence of short circuit currents, the so called shunts [15].

Therefore, it is of great importance to reduce the concentration of C and N in the ingot and also its precipitation (SiC, Si₃N₄).

In this contribution the behaviour of the impurities C and N during the solidification of mc silicon is studied in dependence of its concentration in the feedstock and in dependence of different process conditions. An important source for contamination of the silicon melt by C is via carbon-monoxide in the gas atmosphere [6]. Other C sources could be the silicon feedstock and the crucible coating [7], [8]. The main source of nitrogen contamination is the Si_3N_4 -coating of the SiO_2 -crucible [7], [8].

In this paper results of a study will be shown, where silicon feedstock was used which was intentionally highly contaminated with C and N by adding defined quantities of carbon and nitrogen to the silicon melt. This material simulates a low quality silicon raw material with high carbon and/or nitrogen contamination.

It will be demonstrated that melt convection plays an important role for the C, N distribution in the melt and for the SiC and Si_3N_4 formation in the melt, respectively, and how these defects can be strongly reduced.

Section snippets

Experiments and characterization

Mc-silicon ingots with a diameter of 6cm, length of 4–5cm and a mass of about 0.25–0.3 kg were grown in a laboratory scale vertical-gradient-freeze (VGF) facility which is described in detail elsewhere [8], [12]. The experiments were carried out under two different process conditions I and II. Process condition I is characterized by an open system with conditions of moderate convective transport in the melt. Under these conditions C incorporation via the gas phase will typically occur. For this...

C distribution

Fig. 1 illustrates the measured (FTIR) axial carbon distribution in 10^{17} atoms/cm³ in the centre of a solid silicon ingot for the two different process conditions I and II if uncontaminated pure (EG) silicon feedstock is used. The carbon concentration at the beginning of crystal growth is for process condition I around 4×10^{17} atoms/cm³ and for process condition II two orders of magnitude lower at 4×10^{15} atoms/cm³. The carbon concentration increases rapidly for process I to 10×10^{17} atoms/cm³ already ...

Incorporation of C and SiC formation

From the results of Fig. 1, Fig. 5 it can be concluded that under the poor mixing conditions of process condition I the silicon melt is already saturated with C at the beginning of solidification. This is confirmed by the measured carbon concentration in the silicon ingot at the beginning of the grown crystal i.e. at the crucible bottom for process condition I which is between 3 and 4×10^{17} atoms/cm³ (compare Fig. 1). It is also confirmed by the occurrence of SiC precipitates at the ingot bottom...

Conclusion

The experimental results show that mc silicon ingots completely free of SiC precipitates can be grown if an additional contamination (e.g. from the gas atmosphere or further carbon sources) to the high quality silicon feedstock does not exist. Hence, it is possible to grow mc silicon ingots with carbon content lower than 1×10^{17} atoms/cm³ for 90% of solidified fraction with a distribution following the Scheil equation.

The use of a Si₃N₄ coating of the fused silica crucible wall seems to be...

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References (24)

M. Kittler *et al.*

Microelectronic Engineering (2003)

G. Du *et al.*

Solar Energy Materials & Solar Cells. (2008)

T. Buonassisi *et al.*

Journal of Crystal Growth (2006)

V. Hoffmann, K. Petter, J. Djordjevic-Reiss, E. Enebak, J.T. Håkedal, R. Tronstad, T. Vlasenko, I. Buchovskaja, S....

G. Neuroth, A. Müller, J. Seidel, in: Proceedings of the 21st European Photovoltaic Solar Energy Conference, Dresden,...

J. Chen *et al.*

Journal of Applied Physics (2004)

T. Buonassisi *et al.*

Progress in Photovoltaics: Research and Applications (2006)

F. Schmid *et al.*

Journal Electrochemical Society (1979)

C. Reimann *et al.*, Journal of Crystal Growth (2010),...

C. Reimann, Ph.D.-thesis, in...

K. Søliland *et al.*

Materials Science in Semiconductor Processing (2004)

A. Lawerenz *et al.*

Solid State Phenomena (2004)

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