Magnetic flow control in growth and casting of photovoltaic silicon: Numerical and experimental results

A Poklad1, J Pal2, V Galindo2, I Grants2, V Heinze1, D Meier1, O Pätzold1, M Stelter2, G Gerbeth2

1 Institute of Nonferrous Metallurgy and Purest Materials, TU Bergakademie Freiberg, Leipziger Str. 34, 09599 Freiberg, Germany
2 Institute of Fluid Dynamics, Helmholtz-Zentrum Dresden - Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany
g.gerbeth@hzdr.de

Abstract. A novel, vertical Bridgman-type technique for growing multi-crystalline silicon ingots in an induction furnace is described. In contrast to conventional growth, a modified setup with a cone-shaped crucible and susceptor is used. A detailed numerical simulation of the setup is presented. It includes a global thermal simulation of the furnace and a local simulation of the melt, which aims at the influence of the melt flow on the temperature and concentration fields. Furthermore, seeded growth of cone-shaped Si ingots using either a monocrystalline seed or a seed layer formed by pieces of poly-Si is demonstrated and compared to growth without seeds. The influences of the seed material on the grain structure and the dislocation density of the ingots are discussed. The second part addresses model experiments for the Czochralski technique using the room temperature liquid metal GaInSn. The studies were focused on the influence of a rotating and a horizontally static magnetic field on the melt flow and the related heat transport in crucibles being heated from bottom and/or side, and cooled by a crystal model covering about 1/3 of the upper melt surface.

1. Introduction

Multi-crystalline silicon (mc-Si) is still an important material for the production of solar cells [1,2]. However, the efficiency of the cells is limited by structural defects in mc-Si wafers, such as grain boundaries and dislocations. An improvement of the structural quality of the material is therefore a great challenge for crystal growers. In case of vertical Bridgman-grown mc-Si ingots, so called seeded or seed-assisted growth methods are ascribed a promising potential to address this challenge. Several approaches for seeding are currently under investigation, such as the use of monocrystalline seed plates for a growth of quasi-mono (QM) ingots, firstly described in [3], and the use of a seed layer of polycrystalline Si pieces for a growth of high-performance multi (HPM) ingots [4].

Recently, a novel setup for a Bridgman-type growth of mc-Si ingots in an induction furnace was presented [5], which is characterized by the use of a SiO2 crucible and graphite susceptor with cone-shaped bottom parts. In this paper, a numerical study on the heat and mass transport in this setup is presented, and the growth of conical ingots using the abovementioned seeding methods is demonstrated.

The Czochralski (Cz) technique is the most advanced crystal growth technique. About 95% of the worldwide electronic grade silicon production is based on it, and meanwhile also photovoltaic silicon wafer produced by the Cz method dominate the market compared to the ingot solidification of mc-Si.
For the stabilization and optimization of the Cz growth process externally applied magnetic fields were investigated during past decades [6,7]. This covers static (DC) magnetic fields and alternating (AC) ones such as rotating (RMF) or travelling (TMF) magnetic fields. It is typically attributed to DC fields that they slow-down the flow and damp related turbulent fluctuations, whereas AC fields allow an active generation of a variety of flow structures. A main benefit of AC fields consists in their typically much lower energy consumption compared to DC fields.

A cylindrical Rayleigh-Bénard (RB) cell represents likely the simplest model of the Cz process. It consists of a heated bottom, a cooled upper surface and an adiabatic side wall. For this RB case it was shown that an RMF of a few Millitesla in strength may delay the RB instability and suppress turbulent temperature fluctuations significantly. The latter is due to a transition of the flow structure from a buoyancy driven large-amplitude low-frequency flow structure to a low-amplitude high-frequency flow field if the RMF driven flow dominates [8]. The reduction of the turbulent temperature fluctuations may enable starting the Cz process with a higher initial melt level compared to the standards of today. Besides the RMF, horizontal DC magnetic fields are in industrial use for the Cz process and are known to provide a well-controllable low level of oxygen in the final Si wafer. For the rotating RB case the basic flow field and its linear stability were numerically analysed in [9]. If the bulk flow is dominated by the DC magnetic field then the instability has the form of field aligned convection cells.

For both cases, the RMF and the horizontal DC magnetic field, we report in the following recent experimental results for a physical modelling of the melt flow and the related temperature field which is more close to the real Cz process compared to the RB case.

2. Seed-assisted growth of multi-crystalline silicon in a cone-shaped setup

2.1 Numerical modelling

Numerical modelling of the cone-shaped setup is focused on calculations of the melt flow, as well as the temperature and concentration fields in the melt. It bases on the following programs:

i) CrysMAS: The commercial software package CrysMAS is designed to solve global heat transport phenomena in high-temperature furnaces with complex, axisymmetric geometries [10]. It includes induction and resistance heating, nonlinear heat conduction and radiative heat transfer, which is treated with the method of view factors. In this paper, the program is used for a global thermal modelling of the induction furnace to figure out realistic thermal boundary conditions for a local simulation of the melt.

ii) OPERA 3D: OPERA 3D (© Cobham plc) is a commercial finite element software package for solving three-dimensional electromagnetic simulations on the basis of Maxwell’s equations. The simulation tools are particularly suited for modelling the electromagnetic fields and related physical phenomena in complex experimental setups with an induction coil and electrically conducting components. In this paper, Opera is used to compute the induced current and Lorentz force in the induction furnace, which are needed for a simulation of the Joule heating effect and the electromagnetically driven flow, respectively.

iii) OpenFOAM: OpenFOAM is a free, open source finite volume software for computational fluid dynamics (CFD). It is used for a local simulation of the melt flow, as well as the temperature and concentration fields in the melt in a similar manner as it was done in [11].

Figure 1 shows the CrysMAS model of the central part of the induction furnace with a cone-shaped graphite setup consisting of a conical SiO₂ crucible and a graphite susceptor with a conical inner bottom face. The calculated temperature field results from a global thermal simulation with a certain power of the induction coil and T = 300°C at the outer walls of the furnace. From such calculations, the temperatures at the inner and outer crucible walls were taken to formulate realistic heat flux boundary conditions for a subsequent local modelling of physical phenomena in the melt. The corresponding heat flux profiles at the melt surface and the inner crucible walls are also given in figure 1.
Figure 1. Left: Cross-sectional view of the conical growth setup including a temperature field resulting from a global thermal simulation of the induction furnace. Right: Heat flux profiles (dots – numerical values, lines – adjusted profiles) taken as boundary conditions for a local modelling of the melt (see figure 2).

In figure 2 results of a local modelling of the melt in the conical crucible are presented. Figure 2a shows the Lorentz force density \( f_L = \langle j \times B \rangle / \rho \) induced by the induction coil. The melt flow and the temperature field are shown in figures 2b and c. They were computed by solving the coupled system of Navier-Stokes and energy equations in Boussinesq approximation including Lorentz force and Joule heating terms, which were transferred from the OPERA simulation of the induction furnace:

\[
\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla p + \nu \nabla^2 \mathbf{u} - \beta (T - T_0) \mathbf{g} + f_L
\]

\[
\rho c_p \left( \frac{\partial T}{\partial t} + (\mathbf{u} \cdot \nabla) T \right) = k \nabla^2 T + \langle j^2 \rangle / \sigma
\]
The brackets \(< \cdots >\) in the expression for the Lorentz force density \(f_L\) and in equation (2) specify mean quantities averaged over one AC period. The boundary condition for the flow field at the solid container walls is the no-slip condition \(\mathbf{u} = 0\). For the melt surface either \(\mathbf{u} = \mathbf{0}\) or the conditions for a stress-free, non-deformable surface \(u_n = 0\) and \(\partial u_t / \partial z = 0\) are applied, depending on whether the melt flow is evaluated in an open or enclosed container. As mentioned above, the boundary conditions for the temperature field were taken from a global simulation with CrysMAS in form of given normal heat flux profiles \(-k \mathbf{n} \cdot \nabla T\) at the edges of the melt (see figure 1). Here \(\nu\) is the kinematic viscosity, \(\beta\) is the volume expansion coefficient, \(\mathbf{g}\) is the gravity vector, \(\rho\) is the density, \(c_p\) is the specific heat capacity, \(k\) is the thermal conductivity and \(\sigma\) is the electrical conductivity.

**Figure 2.** Local simulation of the melt in the induction furnace with the conical growth setup for a coil current and frequency of \(I_{\text{eff}} = 260\) A and \(f = 6.3\) kHz, respectively: (a) mass density related Lorentz force density, (b) melt flow, (c) temperature field relative to the melting temperature, and (d) carbon concentration for a growth rate of 20 mm/h, an initial concentration of \(5 \times 10^{-12}\) m\(^{-3}\), a diffusion coefficient of carbon in liquid silicon of \(2 \times 10^{-8}\) m\(^2\)/s, and a partition coefficient of carbon in silicon of 0.07.

The simulation of the concentration field (figure 2d) aims at the distribution of impurities in the silicon melt. For a graphite setup, which is characterized by the use of a graphite susceptor and graphite-based insulation material, carbon is the most important impurity [12,13]. Prior to solidification, the concentration of carbon in the melt is at about the solubility limit of \(5 \times 10^{-12}\) m\(^3\). Therefore, the focus
of simulation was on the concentration at the beginning of the directional solidification process, when
the position of the solid-liquid interface is in the vicinity of the cone tip. For the calculation of the
carbon concentration, we solved the diffusion equation for the species transport:

\[
\frac{\partial c}{\partial t} + (\mathbf{u} \cdot \nabla) c = D \nabla^2 c
\]  

(3)

At the interface, which is assumed to be an ideally flat surface intersecting the cone tip (see figure 2d),
the segregation boundary condition \(- D \frac{\partial c}{\partial n} = u_C c (1 - k_0)\) is used, where \(D\) is the diffusion
coefficient, \(u_C\) is the growth rate and \(k_0\) is the equilibrium partition coefficient.

The following conclusions can be drawn from the results shown in figure 2:

i) The melt flow is mainly driven by the Lorentz force of the induction coil (figure 2a). On the
other hand, the buoyant flow is much less intense by more than one order of magnitude.

ii) The flow velocity in the cone region of the crucible is much lower than in the cylindrical part
(figure 2b) leading to a limited convective heat and mass transfer in this region.

iii) There is a significant axial temperature gradient in the conical part of the crucible (figure 2c),
which favors the control of the crystal-melt interface.

iv) The isotherms near the cone tip are almost flat (figure 2c). This means, the radial temperature
gradient, which is associated with local thermal stresses, tends to zero.

v) The carbon concentration at the beginning of solidification reveals a significant
supersaturation at the assumed interface near the cone tip (figure 2d). This is the consequence
of the low solubility of carbon in solid silicon (partition coefficient 0.07) and the poorly mixed
melt due to the low flow velocity near the cone tip. In a real growth experiment, this would
result in the formation of SiC inclusions, as was indeed observed [5].

2.2 Growth experiments
Si ingots with a cone angle/height of about 120°/25 mm and diameter/height of the cylindrical part
of 105/45 mm were produced with different seeding methods. High-performance multi (HPM) and quasi-
mono (QM) crystals were grown using conventional, polycrystalline Si chunks as feedstock material.
In the case of HPM growth, the crucible was entirely filled with polyc-Si, whereas a single,
monocrystalline Si cone was placed in the conical part of the crucible for the growth of QM ingots. In
a growth experiment, the feedstock was first melted from above, until the position of the melt-crystal
interface was in the conical part of the crucible. The remaining solid silicon formed the seeds for
subsequent directional solidification processes. So the solidification of an ingot started either from a
poly-Si seed layer or from a monocrystalline Si seed resulting in HPM or QM ingots, respectively.
Vertical and horizontal slices were cut from the grown crystals. The slices were polished and etched in
KOH or Secco [14] solutions to reveal the grain structure and dislocation etch pits, respectively. The
etch pit density (epd) was detected by optical microscopy.

The experimental results are shown in figure 3, where HPM and QM crystals are compared with a
conical crystal grown without seed. They can be summarized and discussed as follows:

i) Seed-assisted growth of Si ingots in the conical setup works well. A stable seeding is possible
in the conical part of the crucible, because of the significant axial temperature gradient.

ii) Solidification from a seed layer of poly-Si chunks results in an ingot with a fine grain structure
and a low dislocation density, mainly below the detection limit of \(10^7\) cm\(^{-2}\) (see figures 3c,d
middle). Compared with conventionally grown mc-Si (figure 3d right), the dislocation density
is significantly reduced. This is attributed to an increased ratio of random-type grain
boundaries, which prevent the formation of extended dislocation clusters [4].

iii) The solidification from a cone-shaped Si monocrystal gives an ingot with a large
monocrystalline area, which shows also a low dislocation density (see figures 3c,d left). This
is in contrast to results on QM ingots grown in a cylindrical setup from a set of seed plates,
where pronounced dislocation clusters originating from the joints between seed plates were
found [15]. Hence, with respect to the dislocation density of QM ingots, the use of a single Si seed would be preferable.

iv) Extended polycrystalline areas are detected near the edges of the QM ingot (see figures 3b,c left). Obviously, polycrystalline growth starts at the same time as the solidification process, because first grains appear in the vicinity of the melt – crystal – crucible triple point of the initial seeding interface. On solidification, the grains spread toward the central area of the ingot, indicating a concave shape of the interface and radially outward heat flux. These results show the challenges of the growth of QM ingots with the conical setup. There is a need for further improvements of the seeding process and the temperature field to reduce or even avoid polycrystalline growth.

Figure 3. (a) Scheme of conical Si ingots grown with a monocrystalline seed (left), with a poly-seed layer (middle), and without seed (right). Grain structure of (b) vertical and (c) horizontal slices. (d) Etch pit density of horizontal slices.

3. Physical modelling of the Cz process

3.1 Experiments with an RMF

The modelling experiments for the Cz process followed an approach of increasing complexity. Starting with the generic RB case [8], a modified RB case was then considered with an upper cooled wall smaller than the melt cross-section modelling the solidified crystal [16]. As model melts always GaInSn or pure Ga were used as it is almost standard today [17,18]. Next, a set-up with a realistically shaped, double-walled glass crucible, flown through by a heating fluid, and a water cooled crystal model was installed, both differentially rotatable [19]. Eventually a surface cooling system was added in order to provide realistic heat transfer conditions at the upper free surface of
the melt [18]. This model set-up reproduces in a reasonable approximation the realistic flow and heat transfer conditions in a silicon Cz crucible of limited size. Figure 4 shows two photos and a scheme of this experiment and its components. Main idea behind this type of physical modelling is that the model case allows detailed local measurements of the velocity field and the temperature distribution, which is an essential basis for code validations.

![Scheme of the crucible, crystal model and surface cooler](left). Photo of the double-walled glass crucible (middle). Photo of the set-up with the GaInSn surface, the surface cooler and the rotatable water-cooled crystal model (right).

**Figure 4.** Scheme of the crucible, crystal model and surface cooler (left). Photo of the double-walled glass crucible (middle). Photo of the set-up with the GaInSn surface, the surface cooler and the rotatable water-cooled crystal model (right).

The use of an RMF was investigated mainly because it is considered to be an attractive tool for a significant reduction of temperature fluctuations, which would allow higher initial melt levels in the Cz process, thus increasing the yield of the growth process. Previous experiments for the RB case have shown such strong reductions of the turbulent temperature fluctuations very convincingly [8,16].

The results published in [18,19] examined the suppression of the temperature fluctuations close to the edge of the model crystal under the action of an RMF and/or the rotation of the crystal model. Stirring with an RMF as well as rotating the crystal model causes large-scale temperature oscillations since the single buoyant convection roll is driven around the crucible. Such strong oscillations are an adverse condition in Cz crystal growth. If the field strength of the RMF is increased the convection roll rotates azimuthally faster, and consequently the frequency of the oscillation increases, until the temperature fluctuations lose their periodicity. This transition from a large-scale low-frequency turbulent flow to a small-scale high-frequency turbulent flow was accompanied by a significant (ca. factor 30…50) reduction of temperature fluctuations in the RB and modified RB cases. In the more realistic Cz model as shown in figure 4, some reduction of temperature fluctuations was found as well but by far not as large as in the RB or modified RB cases. The interpretation is that the RMF is still able to break the large-scale thermal structures, but the heating from the side-wall in the Cz case gives rise to a hot jet in the direction of the crystal, which forms a region with a large temperature drop over a small distance. As result, only some reduction of temperature fluctuations is obtained at the rim of the crystal.

The investigations in [18,19] were limited to a fixed aspect ratio of \( a = H/D = 0.59 \) where \( H \) denotes the melt filling height and \( D \) the diameter of the crucible. This is, on one hand, slightly below the value at which in a related RB case a non-axisymmetric single roll appears but, on the other hand, distinctly higher than the filling levels used today in industrial Cz processes. It would be interesting to see whether the above described limited reduction of turbulent temperature fluctuations becomes more pronounced if the aspect ratio is further reduced.
3.2 Experiments with a horizontal DC magnetic field
As mentioned in the introduction, horizontal DC magnetic fields are in use at industrial Cz growth processes as they allow to control the oxygen concentration in a wide range. Their use is somehow surprising since they obviously destroy any axisymmetry, and the details of the flow field which finally gives rise to the oxygen control is not fully understood. Therefore, a series of model experiments has been performed addressing exactly such questions. The set-up under consideration (above named as the modified RB case) used a cylindrical cell as shown in figure 5. With the rotatable crystal model as in figure 4 it was placed in a horizontal DC magnet of a maximum flux density of 326 mT as shown in figure 5.

![Scheme of the cylindrical cell (a) with 1 – liquid metal, 2 – copper bottom plate, 3 – glass wall, 4 – water cooled crystal model made of copper, 5 – surface coolant medium, 6 – cooler spiral. Top view photo of the experiment (b) with red pole faces of the horizontal DC field.](image)

Figure 5. Scheme of the cylindrical cell (a) with 1 – liquid metal, 2 – copper bottom plate, 3 – glass wall, 4 – water cooled crystal model made of copper, 5 – surface coolant medium, 6 – cooler spiral. Top view photo of the experiment (b) with red pole faces of the horizontal DC field.

In [20] the heat transfer, the temperature azimuthal non-uniformity and the onset of oscillations were investigated in detail. It is observed that under certain conditions the integral heat flux may decrease with the magnetic field strength at the same time as the flow velocity increases. The horizontal magnetic field causes a significant azimuthal variation of the melt temperature near the crystal rim. This non-uniformity is explained by radial flow jets in the field perpendicular direction. The jets may be nearly suppressed by fine tuning of the crystal rotation rate resulting in an improvement of the temperature uniformity. The practical realization of such a fine tuning in a real Cz growth is, however, challenging and may be only possible under real-time monitoring of the melt surface temperature. The oscillatory onset in the Cz model with the horizontal DC magnetic field is much delayed in comparison to that in a rectangular RB cell. The difference may be attributed to an an “anchor” in in the form of a fixed cold spot that prevents the field perpendicular movement of rolls. The critical temperature difference increases approximately proportionally to the magnetic field strength in the considered parameter range. The onset conditions are little influenced by the crystal rotation although the basic flow varies significantly. The melt flow may be prone to hysteresis and structural instability leading to loss of symmetry and repeatability under certain conditions.

4. Summary
Numerical and experimental results on the growth of Si ingots in a cone-shaped setup were presented. The melt flow is found to be mainly driven by the Lorentz force of the induction coil. The temperature field is characterized by a significant axial gradient in the conical part of the melt.
With respect to impurities, such as carbon, the melt is poorly mixed near the cone tip. This leads to a local supersaturation of carbon on solidification, which can explain the formation of SiC inclusions observed in other studies. Seed-assisted growth to produce QM and HPM Si ingots is demonstrated. A stable seeding with the initial interface in the conical part of the crucible is possible, because of the high axial temperature gradient. The dislocation densities in QM and HPM ingots are found to be significantly lower than in conventional mc-Si grown without a seed.

For the Cz growth process a unique set of model experiments has been developed, which allows an almost complete measurement of local velocities and local temperatures in the GaInSn melt. Experiments with systematic parameter variations were performed focusing on the influence of either an RMF or a horizontal DC magnetic field on the melt flow and the related heat transfer. A closer comparison with numerical simulations would be highly desirable for future activities with the available Cz model experiments.

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References
[1] Yu H J J, Popiolek N and Geoffron P 2014 Prog. Photovolt. Res. Appl. 24 458
[2] 7th edition of the International Technology Roadmap for Photovoltaic (IPRPV) 2016 (http://www.itrpv.net)
[3] Stoddard N, Bei, W, Witting, I, Wagener M, Park Y, Rozgonyi G and Clark R 2008 Sol. State Phenom. 131-133 I
[4] Yang Y M, Yu A, Hsu B, Yang A and Lan C W 2015 Progress in Photovoltaics: Research and Applications 23 340
[5] Schmid E, Poklad A, Heinze V, Meier, D, Pätzold O and Stelter M 2015 J. Cryst. Growth 416 1
[6] Tomzig E, Virbulis J, von Ammon W, Gelfgat Y and Gorbunov L 2003, Mat. Sci. Semicond. Process 5 347
[7] Vizman D 2015, Handbook of Crystal Growth, vol IIA, ed P. Rudolph (Amsterdam: Elsevier) p 909
[8] Grants I and Gerbeth G 2012 Phys. Fluids 24 024103
[9] Grants I and Gerbeth G 2012 J. Cryst. Growth 358 43
[10] Fainberg J, Vizman D, Friedrich J and Müller G 2007 J. Cryst. Growth 303 124
[11] Galindo V, Niemietz K, Pätzold O and Gerbeth G 2012 J. Cryst. Growth 360 30
[12] Raabe L, Pätzold O, Kupka I, Ehrig J, Würzner S and Stelter M 2011 J. Cryst. Growth 318 234
[13] Funke C, Schmid E, Gärtner G, Reißwenuber S, Füterer W, Poklad A, Raabe L, Pätzold O and Stelter M 2014 J. Cryst. Growth 401 732
[14] D’Secco Aragona F 1972 J. Electrochem. Soc. 119 948
[15] Trempa M, Reimann C, Friedrich J, Müller G, Krause A, Sylla L and Richter T 2014 J. Cryst. Growth 405 131
[16] Cramer A, Pal J and Gerbeth G 2013 Eur. Phys. J. Special Topics 220 259
[17] Dadzis K, Niemietz K, Pätzold O, Wunderwald U and Friedrich J 2013 J. Cryst. Growth 372 145
[18] Pal J, Cramer A, Grants I, Eckert S and Gerbeth G 2015 J. Cryst. Growth 432 69
[19] Cramer A, Pal J and Gerbeth G 2014 Flow Meas. Instrum. 37 99
[20] Grants I, Pal J and Gerbeth G 2017 J. Cryst. Growth 470 58