COST EFFECTIVE GROWTH OF SILICON MONO INGOTS BY THE APPLICATION OF ACTIVE CRYSTAL COOLING IN COMBINATION WITH LARGE MELT VOLUMES IN CZ-PULLER

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ABSTRACT: In the PV industry, there is still a sustained cost pressure in the entire production chain of PV modules. Incidentally, the demand for ever higher efficiencies is another challenge to the material quality in substrate production on the one hand and cell technology on the other hand. For the production of monocrystalline silicon ingots by the Cz process, this means the demand for an extremely productive technology of crystal pulling systems as well as for cost-saving processes. Based on these considerations, a mobile recharge system was developed that allows for the multipulling technique that is the sequential pulling of several crystals from a single crucible. This mobile feeding system offers the unique possibility to serve several Cz-pullers time delayed, which means less investment. In addition, an active crystal cooling device has been developed, which can be retrofitted and allows a significant increase in the pull speed. The combination of active crystal cooling with multipulling enables a strong reduction in production costs of silicon crystals. Another way to reduce costs is to use larger melt weights (primary loadings) to minimize unproductive crystal growing process times (neck, shoulder, cone phase). When increasing the primary loadings, however, the process stability must be taken into account. This raises the question to what extent large melt volumes with the resulting turbulent convection phenomena can be controlled without the use of magnetic fields to dampen the convection phenomena.

Keywords: c-Si, active crystal cooling, multipulling, growth rate, hydrodynamic numbers

1 INTRODUCTION

To meet the demand for an increase of productivity and a reduction of crystallization costs, it is inevitable to optimize the process time as well as the amount of polycrystalline feedstock converted to usable material per process cycle.

During previous conferences we reported the possibility of the average pull speed up to 1.8 mm/min in the body phase for 8-inch ingots, pulling several crystals from a single crucible, and combining both measures [2-4]. This paper examines the possibility of reducing the unproductive process phases relative to the body phase by maximizing the charge weight of silicon in the crucible. For this purpose, theoretical investigations of the process conditions in different hotzone configurations (22 inch, 24 inch, 26 inch) combined with crystal growth experiments were carried out. Using numerical simulations, we created stability diagrams for the different crystal growth conditions and compared them with experimental results. From the simulation calculations we determined the relevant hydrodynamic numbers. The maximum crucible loading in these experiments was 300 kg in a 26-inch crucible. The objective of this experiment was to investigate the process stability without the application of a convection-damping magnetic field.

2 EXPERIMENTAL

2.1 Crystal growth equipment

The crystal growth experiments presented in this paper were performed in three different crystal growth configurations (V1, V2, V3) applied in a SC22 and SC24/26 Czochralski-puller from PVA Crystal Growing Systems GmbH (see fig.1). The crystal growth experiments were performed in different hot zones, the most important parameters of which are given in tab.I. All crystals grown in the reported experiments had a diameter of 8 inches.

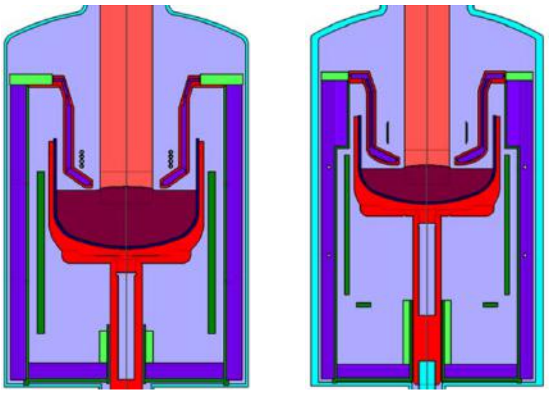

Figure 1: Crystal growth configuration of V1 and V2basic (left) and V2, V3 (right)

growth configuration	Cz-puller type	hotzone	crucible dimension [inch]
V1	SC 22	fig.1 left	22
V2 basic	SC 24	fig.1 left	24
V2	SC 24	fig.1 right	24,26
V3	SC24/26	fig.1 right	26

Table I: Crystal growth configurations

2.2 Hotzone design with and without active crystal cooling

The physical limiting parameter for the growth rate of ingots in a Czochralski configuration is the release of the latent heat of fusion at the interface crystal/melt by heat conduction through the growing crystal and its dissipation by heat radiation over the surface [1]. Due to the design of the hotzone and here in particular of the inner heat shield, the temperature gradient at the phase boundary can be varied within certain limits. In this work an average pull speed of 1.3 mm/min could be achieved only by optimizing the hotzone design. In order to further increase the pull rate, an active crystal cooling device (ACC: Active Crystal Cooling) in the form of a water-cooled spiral was installed within the inner heat shield. The application of the ACC increases the net heat exchange between the crystal surface and the environment, thereby increasing the axial temperature gradient at the phase boundary. The main parameters of the cooling element are the cooling water flow, the vertical distance to the melt surface, the radial distance to the crystal and the emissivity of the cooling element. The crystal growth configuration including the active cooling system is shown in fig.2. Safety considerations and other details of the ACC are described in detail in [2].

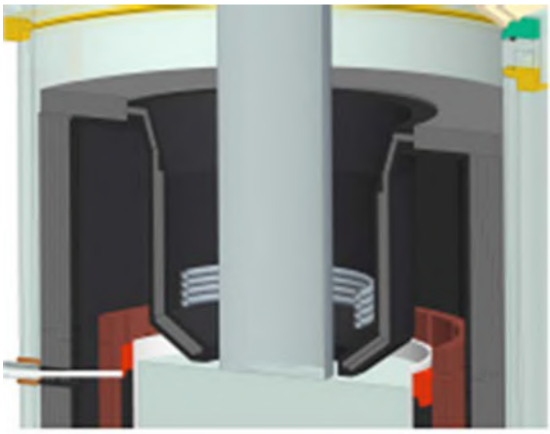

Figure 2: Arrangement of the active crystal cooling device (spiral) in the hotzone

2.3 Mobile recharge system (MRS)

In order to reduce the time consuming temperature cycles between the growth runs and to reduce the consumption of crucibles the multipulling process, in which several crystals are sequentially pulled out of a single crucible, has been established. Between two growth runs, the hot crucible is then refilled with silicon, which can be done in different ways [2]. The PVA Crystal Growing Systems GmbH has developed a mobile recharge system (MRS), which offers the unique possibility to serve several Cz-pullers time-delayed. The advantage of the MRS system is the possibility to perform the feed process while the crystal is cooling in the receiving chamber. Fig.3 shows the MRS system docked on a SC22 Cz-puller. Further considerations on multipulling, such as: impurity redistribution in ingots grown from a replenished melt is given in [3] and an economic analysis of the multipulling process combined with active crystal cooling is published in [4].

2.4 Crystal growth

Three different crystal growth configurations were investigated theoretically and experimentally to evaluate the main key parameters influencing the crystal growth conditions with and without ACC. Fig.1 shows schematic drawings of the hotzones installed in the SC22 and SC24/26 puller. The crucibles used had diameters of

Figure 3: Mobile recharge system docked on a Cz-puller

22, 24 and 26 inches with the corresponding possible melt volumes of 150 kg, 200 kg and 300 kg. The graphite parts had different geometries and partially different insulation material. Corresponding to the crucibles sizes the growth configurations are designated in this paper by the abbreviations V1, V2 and V3. All three arrangements are equipped with an active crystal cooling system (ACC). In the arrangements V2 and V3 a bottom heater can be used. The main growth parameters used in the crystal growth experiments are given in tab.II.

	crystal rotation	crucible rotation	growth rate	bottom heater
V 1	10	-10	1.1 -1.5	no
V 2	10	-6	0.9 -1.8	yes
V 3	10	-6	1.6 -1.8	yes

Table II: Main growth parameters in the different growth configurations

2.5 Numerical simulation

The crystal growth arrangements and the associated process conditions were examined and optimized by means of numerical simulation calculations. For this purpose, the commercially available CGSim program package [1] was used. The heat and mass transfer task was solved in a 2D axisymmetric quasi-steady state approximation. In the gas atmosphere, heat conduction, radiation heat exchange between solid surfaces and gas convection, considered within Navier-Stokes equations,

[1] STR Group, Inc., St. Petersburg, Russia

are simulated. The convective heat transport in the melt is calculated using a one equation-turbulence model, which takes into account buoyancy convection, Marangoni convection and forced convection by crucible and crystal rotations. The released heat of crystallization is also considered in the calculations.

3 RESULTS

3.1 Interface deflection

An important quality criterion of the hotzone design with regard to process stability and material quality is the deflection of the phase boundary in the growing crystal. Ideally, but not feasible, the curvature of the phase boundary melt/crystal should be flat. The parameter of the deflection of the phase boundary in the crystal center H (fig.4) is plotted versus the average pull speed in fig.5 for a crystal length of 500mm and versus the crystal length in fig.6 for hotzone V3 and different pull speeds.

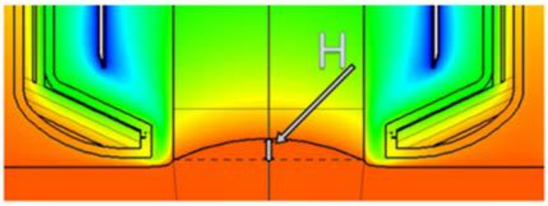

Figure 4: Sketch of the cooling device and the inner heat shield. H indicates the deflection of the phase boundary.

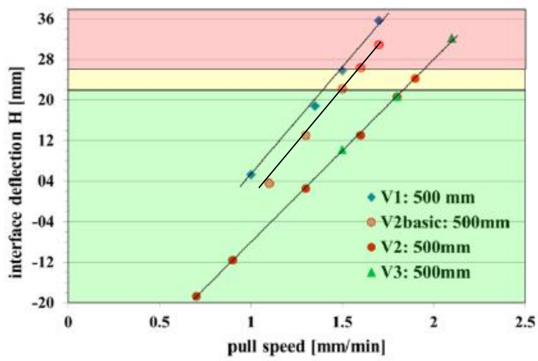

Figure 5: Stability diagram for the different crystal growth configurations

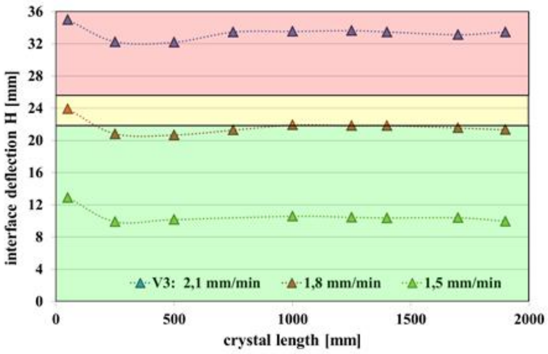

Figure 6: Stability diagram for the different pull speeds in the body phase versus crystal length in V3

In the diagrams three different regions are shown: In the stable growth region (green) the system is insensitive to changes in the average pull speed. This region is robust and suitable for industrial production [1]. In the metastable growth region (yellow) all growth parameters have to be well tuned. Small changes can lead to unstable growth with loss of shape, i.e. spiral growth. In the unstable growth region (red) no regular crystal growth is possible. In the diagram of fig.5 the points represent individual results of the simulation calculations for the various crystal growth configurations (V1-V3). In all hotzone configurations the active crystal cooling (ACC) system was installed. In the configurations V2 and V3, optimized heat insulation as well as an optimized inner heat shield was used. This measure was decisive for the improvement of the growth conditions, which is expressed in a shift of the "stability straight line" from V2basic to V2 towards higher pull speeds. V2basic refers to the heater configuration prior to thermal optimization. Noticeable is the fact that both the 24-inch hotzone V2 and the 26-inch hotzone V3, being nearly the same in geometry, show practically the same H-changes. Fig.6 shows the calculated deflection of the phase boundary for three different pull rates in the growth configuration V3. From a crystal length of about 300 mm, the phase boundary shows an approximately constant deflection. This result was also confirmed experimentally by LPS (Lateral Photovoltage Scanning) measurements [4] on vertically sliced crystal samples. Similar results were obtained for the configurations V1 and V2.

3.2 Hydrodynamic numbers

Dimensionless similarity measures (hydrodynamic numbers) can be used for the characterization of flow states. These hydrodynamic numbers contain specific parameters as well as technological boundary conditions which describe the respective flow conditions in the melt [5]. By these numbers, flow conditions can be qualitatively estimated. Even if these measures are often overrated, they still allow comparison and qualitative assessment of different process conditions. The following dimensionless numbers[2] are used to evaluate the different crystal growth conditions:

$$Ra = \frac{g * \beta * \Delta T * h^3}{\nu * \kappa}$$

The **Rayleigh number Ra** represents the ratio of heat transfer by thermal buoyancy convection to heat conduction.

$$Ta = \frac{4 * \omega^2 * r^5}{\nu^2 * h}$$

The **Taylor number Ta** is a measure of the crucible rotation rate and describes the ratio of rotational force to friction force.

$$Ro = \frac{g * \beta * \Delta T * h}{\omega^2 * r^2}$$

The **Rossby number Ro** indicates the ratio of buoyancy forces to rotational forces.

$$Ma = \frac{\frac{\partial \gamma}{\partial T} * \Delta T * L}{\nu * \kappa * \rho}$$

The **Marangoni number Ma** describes the ratio of the

[2] g: gravitational acceleration, β: thermal expansion coefficient, ΔT: temperature difference, h: characteristic length, ν: kinematic viscosity, κ: coefficient of thermal conductivity, ω: crucible rotation rate, r: crucible radius, ∂γ/∂T: Marangoni parameter (γ: surface tension), ρ: melt density

heat transfer by interfacial convection to the dissipative heat transfer by heat conduction and takes into account the occurring surface effects.

It is generally acknowledged in the literature that silicon melts have turbulent character in growth configurations as considered here [6]. Such flow conditions usually lead to undesirable temperature fluctuations at the phase boundary, degrading the crystal quality. These temperature fluctuations can be influenced in certain limits by means of the crucible rotation. The extent of buoyancy convection f(h) with h melt height versus forced convection f(ω) with ω crucible rotation rate is plotted in fig.7-fig.9 for the different growth configurations.

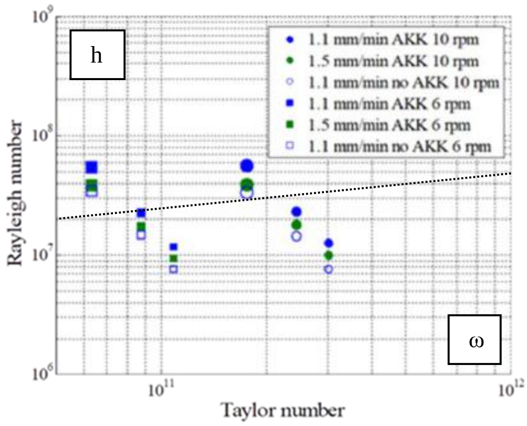

Figure 7: Rayleigh number versus Taylor number in crystal growth configuration V1. The size of the symbols corresponds to the melt quantity (75 kg, 100 kg, 150 kg).

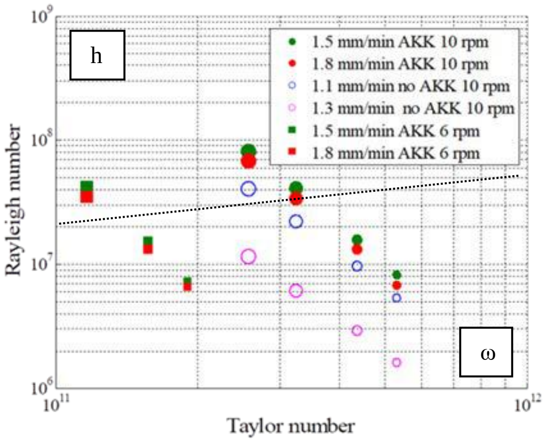

Figure 8: Rayleigh number versus Taylor number in crystal growth configuration V2. The size of the symbols corresponds to the melt quantity (75 kg, 100 kg, 150 kg, 200 kg).

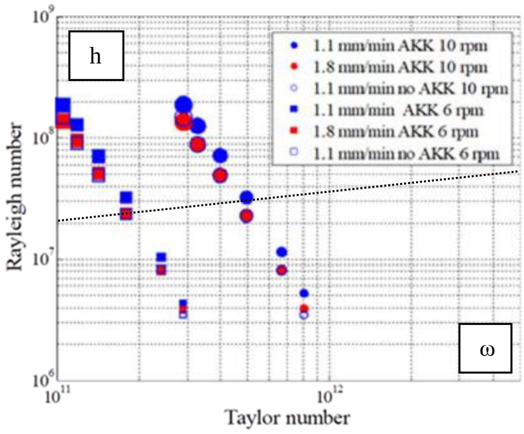

Figure 9: Rayleigh number versus Taylor number in crystal growth configuration V3. The size of the symbols corresponds to the melt quantity (75 kg, 100 kg, 150 kg, 200 kg, 250kg, 300 kg).

In the diagrams, a fictitious boundary - known in the literature as the Rossby criterion - is shown as a dashed line, which is supposed to represent a transition from oscillatory to turbulent flow [7]. However, the plot can only be interpreted as an indication, since it was determined by Rossby [8] in a special Bénard configuration, which can be hardly transferred to the Czochralski configuration. The legends inserted in the diagrams indicate the average pull speed, the crucible rotation [rpm] and the use of the active crystal cooling (AKK). The size of the symbols corresponds to the melt quantity, which can be assigned to a specific aspect ratio h/d (melt height to crucible diameter) of the molten charge. Tab. III shows the corresponding data.

melt volume [kg]	75	100	150	200	250	300
V1: h/d	0.31	0.39	0.54			
V2: h/d	0.26	0.32	0.44	0.56		
V3: h/d	0.24	0.26	0.36	0.45	0.55	0.65

Table III: Aspect ratio h/d for the different growth configurations

Fig.10-fig.12 show the Rossby number plotted versus the Taylor number. Essentially, two parameters determine this hydrodynamic numbers: the crucible rotation and the maximum temperature difference which is driving the buoyancy convection.

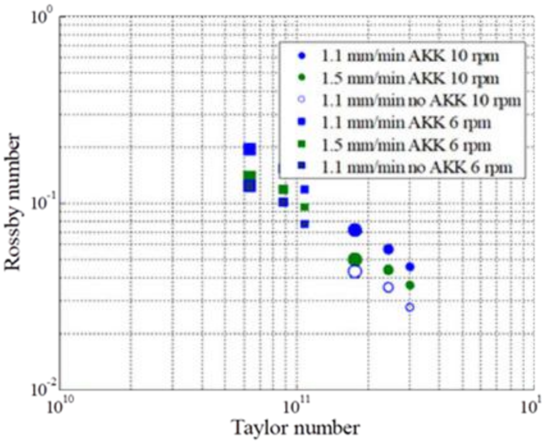

Figure 10: Rossby number versus Taylor number for the crystal growth configuration V1. The size of the symbols corresponds to the melt quantity (75 kg, 100 kg, 150 kg).

In the process, both the buoyancy convection and the forced convection driven by rotations of crystal and crucible clearly outweigh the effects of Marangoni convection. The thermally driven Marangoni convection is a near-surface flow directed towards the edge of the crystal. The Marangoni flow seems to have a stabilizing influence on the three-phase line. Fig.13-fig.15 show the Marangoni numbers calculated for the different crystal growth configurations. The main growth parameters are given in the legend. It becomes clear that the use of active crystal cooling with otherwise identical growth parameters enhances the Marangoni effect. A higher pull speed causes an increased release of heat of crystallization, which can lead to a reduction of the Marangoni convection [1].

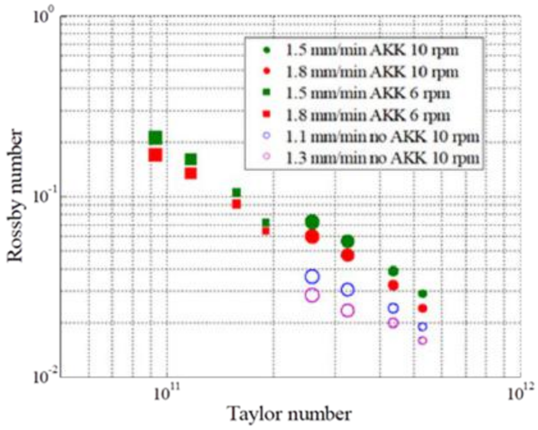

Figure 11: Rossby number versus Taylor number for the crystal growth configuration V2. The size of the symbols corresponds to the melt quantity (75 kg, 100 kg, 150 kg, 200 kg).

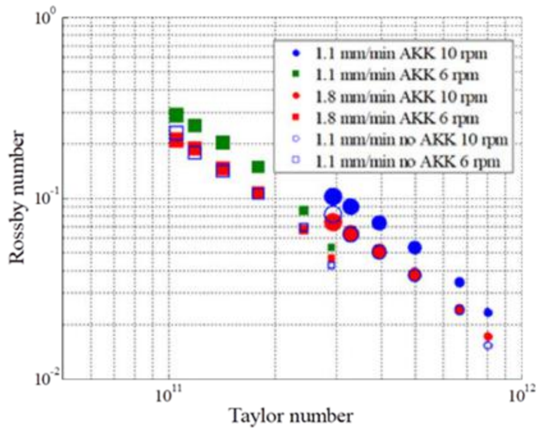

Figure 12: Rossby number versus Taylor number for the crystal growth configuration V3. The size of the symbols corresponds to the melt quantity (75 kg, 100 kg, 150 kg, 200 kg, 250 kg, 300 kg).

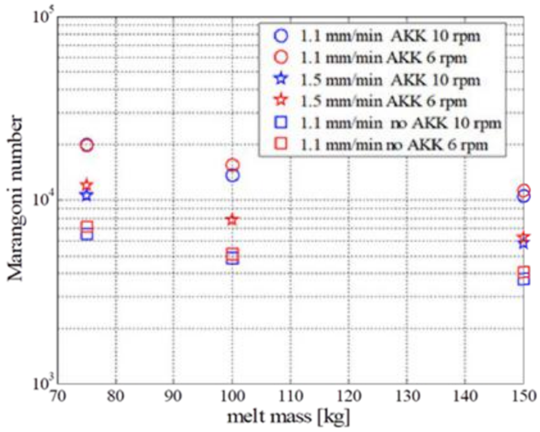

Figure 13: Marangoni number versus melt mass for the crystal growth arrangement V1.

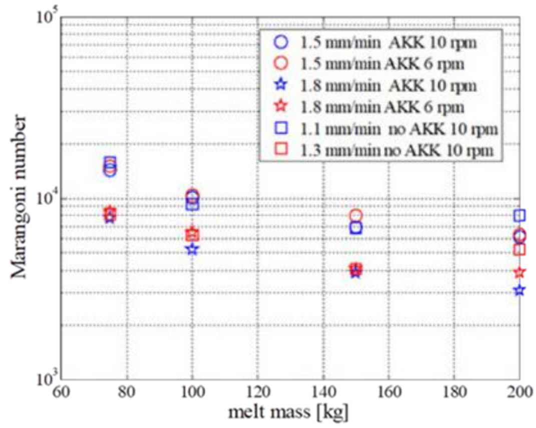

Figure 14: Marangoni number versus melt mass for the crystal growth arrangement V2

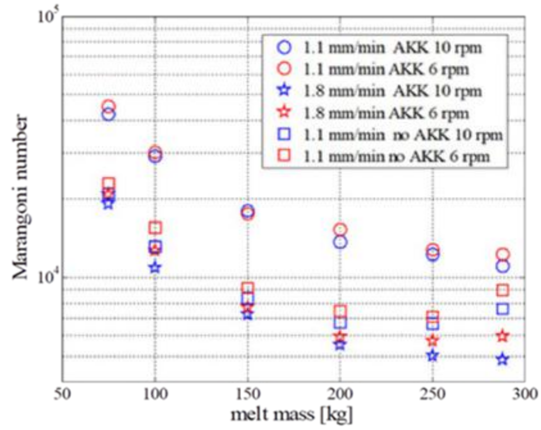

Figure 15: Marangoni number versus melt mass for the crystal growth arrangement V3

4 DISCUSSION

4.1 Theoretical considerations

Fig.16 and fig.17 show hydrodynamic stability diagrams from the literature. It should be noted that both diagrams describe specific model cases that can only be qualitatively transferred to the Czochralski arrangements. Fig.16 describes a stability diagram for rotating double cylinders (radial temperature gradient) with a model fluid [9]. The crystal growth conditions examined here are shown in red dashed lines.

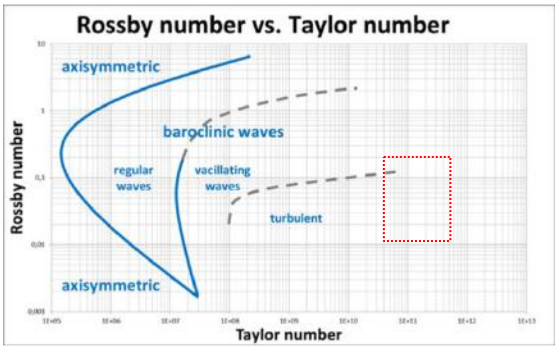

Figure 16: Schematic representation of the convection conditions in a model arrangement [9].

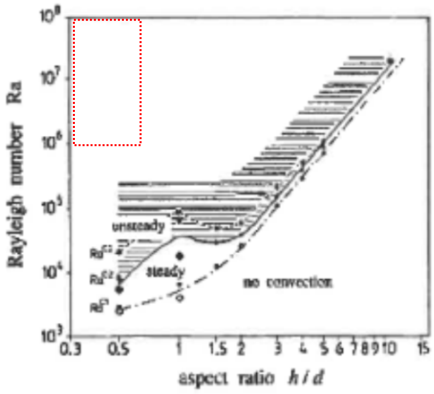

Figure 17: Stability diagram of buoyancy convection in metal and semiconductor melts (Pr~0.01) in a cylindrical model with insulating sidewalls [10]

It is clear from both diagrams that the crystal growth configurations described here are dominated by turbulent melt convection. Convection-inhibiting influences of the vertical crucible walls are negligible in any of the considered and industrially relevant aspect ratios h/d. Experimental and numerical considerations show that with decreasing h/d ratio, and thus during the growth process, the growth conditions become more stable [11]. This fact can be attributed to a change in the flow configuration towards more stable conditions. In addition, the elliptical curvature of the crucible bottom has a dampening effect on the buoyancy convection [12].
The diagrams of the hydrodynamic numbers (fig.7-fig.15) show results with and without active crystal cooling. The comparison of the Rayleigh number with the Taylor number (fig.7-fig.9) shows that the configurations V1, V2 and V3 seem to be in a kind of flow transition. The use of active crystal cooling allows higher pulling speeds, but at the same time leads to slightly increasing buoyancy convection (Rayleigh number). The active crystal cooling causes not only an enhanced heat exchange with the crystal but also with the process environment. Therefore, at the same time more heating power must be supplied to the system, which is expressed in an increased axial temperature gradient.
By adapting the different growth parameters, it may be possible to influence the stability of the convection phenomena within certain limits. A higher rotation rate of the melt should have a stabilizing influence on the flow configuration. With a larger crucible rotation, however, the increase in baroclinic instabilities can be expected. This means that, according to the Rossby-Taylor diagram (fig.16), the growth parameters should be changed to larger Rossby numbers, which can also be achieved by reducing crucible rotation, which is consistent with results in the literature [13]. This fact is not a contradiction but clarifies the reciprocal and extremely sensitive dependence of the growth parameters.
Fig.13-fig.15 show the Marangoni number over the melt quantity. The Marangoni convection has a negligible influence on the flow configuration in the melt volume. However, the Marangoni effect seems to be weakened with increasing pull rate, which can lead to an unstable phase boundary. Exceeding a critical pull rate will result in the loss of cylindrical shape of the grown crystal. Such phenomena are experimentally proven and also reported in the literature [14].

4.2 Crystal growth experiments

Various crystal growth experiments were carried out in the crystal growth configurations V1, V2 and V3. The main growth parameters are shown in tab.II. The main focus of the growth experiment in the growth configuration V3 with a charge of 300 kg was on the feasibility of growing a crystal without the use of a convection-damping magnetic field. From this point of view the fluctuations of the diameter and the average pull speed in the body phase were examined and compared with crystal growth experiments in the other growth configurations. Since larger melt volumes can be expected to increase the temperature fluctuations they were recorded after a stabilization period before neck growth and compared. The results are shown in fig.18, there are barely any differences.

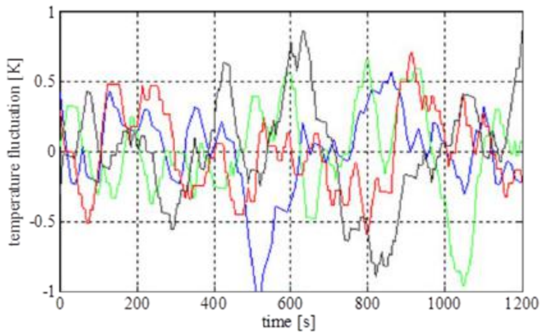

Figure 18: Measured temperature fluctuations on the stabilized melt surface 20 minutes before the start of the neck phase in the different growth configurations: blue line: V1, ACC, 120 kg; green line: V2, ACC, 100 kg; red line: V3, ACC, 185 kg; black line: V3, ACC, 300 kg.

Characteristics for the stability of the growth conditions in the different growth configurations are the diameter fluctuations and the resulting fluctuations in the pull speed. It can be seen from fig.19 and fig.20 that an increase in the pull speed as well as in the loaded charge result in an increase of the fluctuations both in diameter and in the average pull speed. Since a radius fluctuation $\dot{r}_i$ at deviation from the meniscus angle α_i for a constant diameter is proportional to the pull rate v_g, i.e. $\dot{r}_i = v_g tan(\alpha_i)$ [15], a larger diameter fluctuation is to be expected at a high growth rate. This fact, in turn, inevitably leads to a greater variation in the average pull speed since it is the control parameter in the diameter closed control loop. With increasing melt volume (h/d ratio) and increasing pull speed, the fluctuations in crystal diameter and the resulting pull speed increase. This can be counteracted within certain limits by adapting the control loops (PID parameters). In the test arrangement V3 with a weight of 300 kg, the growth parameters are not yet optimized. Nevertheless, it can be stated that the use of large amounts of melt is possible without the use of a magnetic field. However, the growth and control parameters have to be optimized for each process phase. The process windows become increasingly narrow as the melt quantity as well as pull speed are increased.

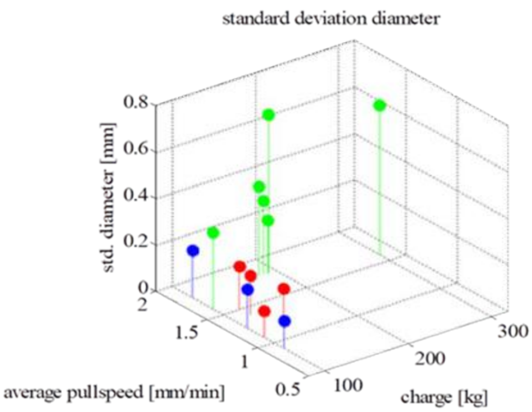

Figure 19: Standard deviation of the diameter versus charge quantity and setpoint of the average pull speed for crystal growth experiments in V1 (blue symbols), V2 (red symbols) and V3 (green symbols) growth configurations.

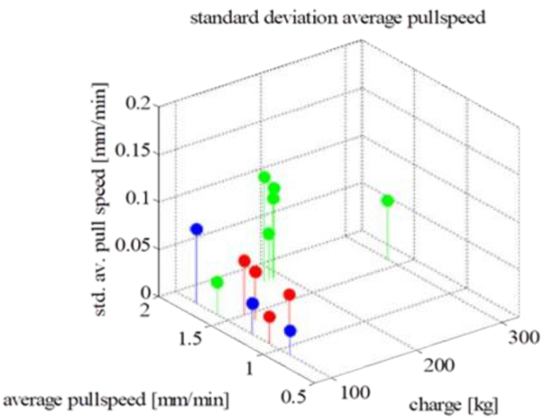

Figure 20: Standard deviation of the average pull speed versus charge quantity and setpoint of the average pull speed for crystal growth experiments in V1 (blue symbols), V2 (red symbols) and V3 (green symbols) growth configurations.

5 CONCLUSIONS

Maximizing the melt volumes in existing Cz-crystal pullers may be an attractive way to further reduce crystallization costs in the short term. From the point of view of material yield, material quality and system handling, the transition to larger Cz-pullers including a magnet seems inevitable in the longer term.

6 ACKNOWLEDGEMENTS

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