Physical model of Czochralski crystal growth in a horizontal magnetic field

J. Pal[1], I. Grants[1,2], S. Eckert, G[1]. Gerbeth[1]

[1]Helmholtz-Zentrum Dresden – Rossendorf, Bautzner Landstr. 400, 01328 Dresden, Germany
[2]Institute of Physics, University of Latvia, Miera iela 32, LV-2169 Salaspils, Latvia

Corresponding author : j.pal@hzdr.de

Abstract
A low temperature liquid metal model of the Czochralski crystal growth process is considered experimentally under conditions of high aspect ratio. In this paper we focus on the influence of a horizontal magnetic field (HMF) on the radial flow field and present first results from related model experiments. The flow is measured by means of the ultrasound Doppler velocimetry (UDV).

Key words: Czochralski Crystal Growth, Horizontal Magnetic Field, Flow Measurements

Introduction

About 95% of the mono-crystalline silicon is produced by the Czochralski (CZ) method. In the process, a single seed crystal is attached to the free surface of molten high purity silicon in a cylindrical quartz crucible. Under rotation and simultaneous pulling of the seed the melt crystallizes and a mono-crystalline rod develops as the final product. This method is now routinely applied for the production of 300 mm diameter single crystals. Further size increase is certainly possible, though there are technical challenges to be solved. To grow a longer 450 mm crystal with the present equipment, a higher initial filling level would be needed. However, the melt instability is increasingly difficult to suppress by crucible/crystal rotation as the height of the melt is increased.
The fluid flow inside the crucible serves as a carrier for mass and temperature and affects crucially the quality of the growing crystal. The horizontal magnetic field (HMF) may improve conditions in the melt during large silicon single crystal growth by the CZ technique. This observation might be counter-intuitive as the HMF evidently breaks the rotational symmetry. A previous study [1] has shown, that the HMF is not able to significantly delay the Rayleigh-Bénard (RB) instability in a rotating cylinder. It has been observed in a large aspect ratio rectangular box that an oscillating flow sets in soon after the linear onset [2].
We have started a longer–term research program to investigate the effect of an HMF on the thermally driven flow within a setup which has some striking similarities with the Czochralski (Cz) silicon crystal growth process. The research focuses on the development of the flow and temperature field under the influence of the asymmetrically applied magnetic field. The results should help for answering various questions, for instance, whether a stabilizing effect of the HMF on the flow can be expected in the Cz process or which consequences arises with respect to the mean heat flux expressed by the Nusselt number *Nu(Ra,Ha)* or regarding the local temperature field near the crystal edge. Besides, the ongoing experimental study is also considered as a benchmark for comparison with numerical codes. Therefore, to serve the latter purpose, the boundary conditions should be well defined. In the present paper we present first preliminary flow measurements from the ongoing measurement campaign.

Basic principles in modelling the Czochralski technique

The simplest model of the CZ facility might be a RB configuration, in particular a cylindrical cell heated from below and cooled at the top which is characterized by a height H and diameter $2R$ and adiabatic insulated side walls. Applying a temperature gradient $T = T_b - T_t$ between the bottom and top sides, buoyant convection occurs when T exceeds some critical value. The dynamics of the mere thermally induced convection may be described by three control parameters. First of all the dimensionless Rayleigh number Ra is the crucial parameter in modelling buoyancy and describes its strength: $Ra = g\ TH^3/$, where is the thermal expansion coefficient , the kinematic viscosity, the thermal diffusivity of the fluid, and g the gravitational acceleration. The second control parameter, the Prandtl number Pr, takes into account the heat transport within the fluid and is given by the ratio of the thickness of viscous and thermal boundary layers: $Pr =$. In general, molten metals and semiconductor melts are low Prandtl number fluids with Pr in the order of 10^{-2}, which means that the heat diffuses quickly in comparison with the convective transport. The third control parameter, the aspect ratio $a = H/(2R)$ concerns the geometry of the setup and affects crucially the developed convective pattern inside the melt. Owing to a, the variation of the convective patterns was studied numerically and experimentally in [3] for Pr 29; a single roll convective motion corresponding to the azimuthal wave number of $m = 1$ was found for

a ≥ 0.63. This flow is often termed wind or large scale circulation (LSC) and is frequently observed in RB systems. More details on such wind can be found in [4] and references therein. For smaller *a* down to about 0.3 the authors of [3] observed $m = 0$, which is a torus and thus axisymmetric. The initial filling level in a real industrial CZ facility does not reach $a = 1$, it is even lower than $a = 0.5$ and decreases continuously during the process. One of the reasons not using high *a* up to 1 in practice might be the generation of non-axisymmetric modes implied by the RB configuration.
The Hartmann number $Ha=BL(\ \ /\ \ )^{1/2}$ occurs as another control parameter if external magnetic fields are exposed to the system. Thereby is *B* the magnetic induction, *L* a typical length scale in the system, and   the electrical conductivity of the melt. The Hartmann number represents a measure for the ratio between the electromagnetic body force and the viscous force.

The experimental setup

The object of the investigation is a modified Rayleigh-Bénard (RB) configuration, a cylindrical melt column of variable aspect ratio homogeneously heated from below. The photo in Fig. 1 illustrates the experimental setup mounted between the HMF producing coils. As working fluid the ternary alloy GaInSn [5] was used because it remains liquid at room temperature and as distinguished from mercury it is non-poisonous. Moreover, its low Prandtl number is similar to that of molten silicon.
The heating was realized by an electrical heating plate embedded in a massive copper disc to achieve isothermal boundary condition. Several thermocouples were installed inside the copper disc for monitoring its temperature. The upper thermal boundary condition in a Czochralski system is accounted for a partially cooled surface. The partial cooling in our experiment covers approximately the same fraction area as the crystal does in an industrial facility. It is realized with a circular heat exchanger (cold finger) mounted concentrically at the top of the experimental cell. The cold finger is optionally rotable. A precise control of the temperature of the cold finger is realized by supplying it with coolant fluid at high flow rate from a thermostat having a large reservoir. The latter is regulated by a PID circuit. The temperature of the cold finger is monitored at various positions.
For the purpose to achieve adiabatic boundary conditions at the side walls a borosilicate glass pipe was chosen as experimental cell owing to its poor heat conductivity. Furthermore, the glass pipe allows the transmission of ultrasound for flow measurements. The drawing in Fig. 2 illustrates the cell and the adjustable holder for the ultrasound sensors. During the measurements, the apparatus was embedded in mineral wool to minimize the lateral heat loss.
In the real Czochralski facility thermal radiation from the free hot silicon melt is a considerable heat sink. A distinctive feature of the present setup is the possibility to model the heat loss from the melt surface. The surface is therefore covered by an electrolyte layer which is cooled by a copper spiral completely immersed into this layer (cf. Fig. 1). Instead of pure water a weak aqueous solution of KOH was used as electrolyte to protect the GaInSn surface from oxidation.
Flow velocities were measured by the UDV technique, the principle of operation is described in the pioneering work of [6]. Mainly two features render UDV predestinated for the present work. Firstly, it works for opaque media including liquid metals. Secondly, it allows the quasi-simultaneous measurement of an entire profile of the local velocity components in direction of the sound propagation along the ultrasonic beam. The readings of the 8 MHz transducers (type TR0805LS, Signal-Processing, Lausanne, Switzerland) were taken by a DOP2000 velocimeter (model 2032 from the same manufacturer).

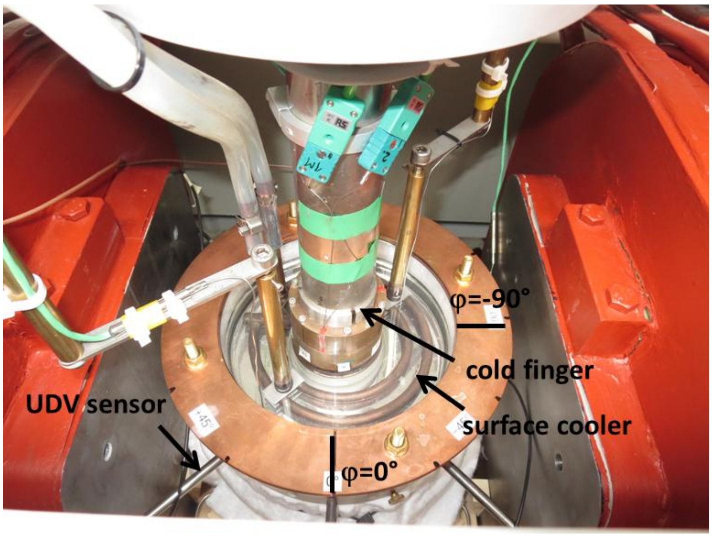
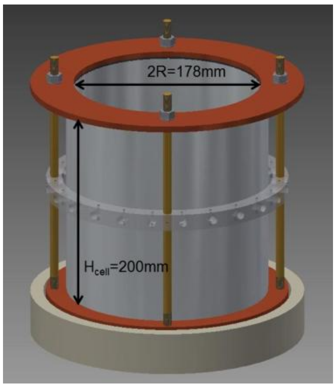


Fig. 1: Photo of the experimental setup mounted inside of the HMF coil system.



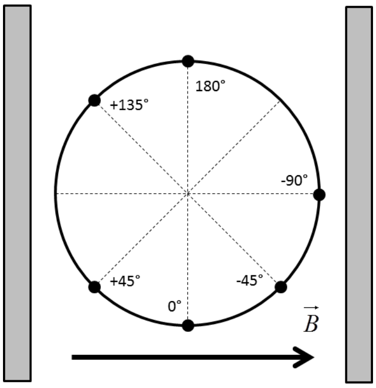
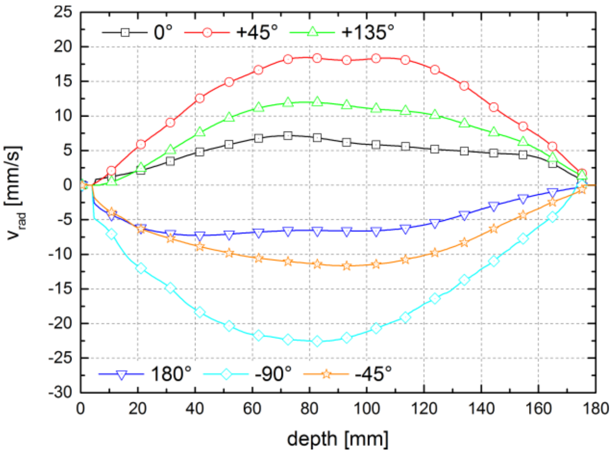


Fig. 2: The cell covering the liquid metal with its dimensions and the UDV sensor holder.

Fig. 3: Azimuthal positions of the UDV sensors where measurements were performed.

Results

All measurements performed for the present paper were done under the same thermal boundary conditions. The bottom heater was always operated with a constant power of 1035 W. After a certain thermalization phase of the experimental cell a constant temperature gradient of T = 41 K was obtained between the heater and the cold finger, which corresponds to the Rayleigh number of Ra = 3.7 10^7. The cell filling height was kept constant at H = 150 mm, which results in an aspect ratio of a=0.84. Hence, a single roll convection cell (wind) is expected. Six UDV sensors were positioned slightly below the liquid metal surface at different azimuthal positions to measure the radial flow velocity across the diameter. The sketch in Fig. 3 indicates these measurement positions. The connecting diameter 0°/180° is aligned perpendicular and the -90°/ +90° one parallel to the direction of the HMF. Fig. 4 shows velocity profiles measured without applied magnetic field. The findings suggest the occurrence of the wind in the meridional plane spanned by the positions -90°/+90°. Hence, the wind comes upward along the side wall to the top at +90°, crosses the cell along the diameter, descends at -90°, and closes along the diameter at the bottom.



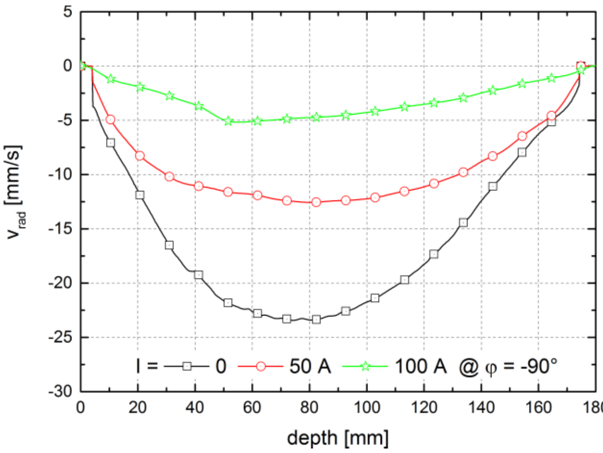


Fig. 4: Radial velocity profiles across the diameter measured at different azimuthal positions. A positive sign of the velocity indicates a flow direction moving away from the sensor.

Fig. 5: Velocity profiles at the azimuthal angle -90° for different feeding currents of the HMF coils.

Fig. 5 shows velocity profiles measured at -90° for the cases without and two different magnetic fields. The indicated coil feeding currents of I = 50/100 A correspond to the magnetic induction B = 29.3/59.5 mT, or Ha = 102/206. Even though the flow is aligned parallel to the HMF a strong damping in the flow velocity occurs, which is at the center (r = 89 mm) about 54% for Ha = 102 and 78% for Ha = 206. Fig. 6 displays the velocity profiles recorded at two other positions for Ha = 206. Firstly at = 0°, where the flow is perpendicular to the HMF, and, secondly at = -45°. A comparable velocity damping of about 83% and 73% occurs for the same Ha number at = -45° and = 0°, respectively. Even a partial change in the flow direction is observed at = 0° by applying a HMF with Ha = 206.

Concerning the flow structure a flow reversal behavior was observed. The large scale recirculation roll (wind) remains thereby as the main hallmark of the flow but it changes completely the flow direction. Such flow reversals for high *Ra* number under high aspect ratio conditions are well known in the literature, [7, 8] to quote a few, and still under investigation. In the measurements done so far flow reversals were observed not only in the mere buoyant case but also for the case when the HMF was switched on. Fig. 7 summarizes measured velocity profiles in this cases.

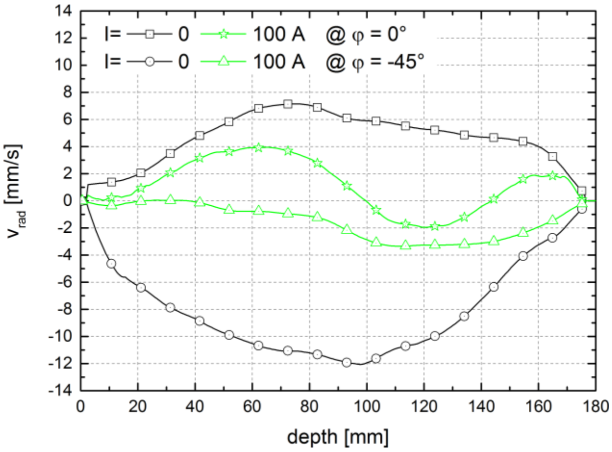


Fig. 6: Velocity profiles at the azimuthal angle 0° where the flow is perpendicular to the HMF, and -45°, which is between the parallel and perpendicular one.

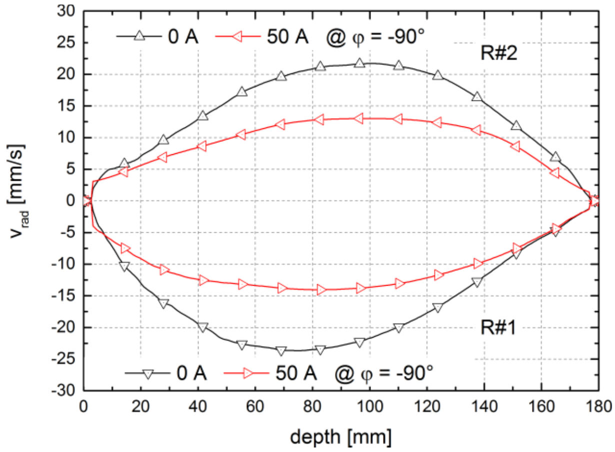


Fig. 7: Velocity profiles measured without (R#1) and with (R#2) occurred flow reversal.

Conclusions and outlook

First flow measurements were performed in a laboratory CZ model using a low Prandtl number melt contained in a volume of high aspect ratio. The thermally driven fluid flow was exposed to a horizontal DC magnetic field. A strong damping of the flow was observed for the flow velocity parallel to the orientation of the HMF. Also flow reversals were observed during the measurements. Belonging to ongoing investigations, additional temperature measurements will be integrated in the vicinity to the rim of the crystal mockup (cold finger). Measurements as function of *Ra*, *Nu*, *Ha*, aspect ratio *a*, and crystal mockup rotation rate are on the way to investigate the questions pointed out in the Introduction and to compare the measurements with numerical simulations.

Acknowledgment

Financial support of this research by the German Helmholtz Association in the frame of the Helmholtz-Alliance LIMTECH is gratefully acknowledged.

References

[1] I. Grants, G. Gerbeth, J. Cryst. Growth 358 (2012), 43-50.
[2] U. Burr, U. Müller, J. Fluid Mech. 453 (2002), 345-370.
[3] F. Hébert, R. Hufschmid, J. Scheel, G. Ahlers, Phys. Rev. E 81 (2001), 046318.
[4] J. Niemela, L. Skrbek, K. Sreenivasan, R. Donnelly, J. Fluid Mech. 449 (2001), 169–178.
[5] Y. Plevachuk, V. Sklyarchuk, S. Eckert, G. Gerbeth, R. Novakovic, J. Chem. Eng. Data 59(3) (2014), 757–763.
[6] Y. Takeda, Nucl. Eng. Des. 126 (1991), 277-284.
[7] K.R. Sreenivasan, A. Bershadskii, J.J. Niemela, Phys. Rev. E 65 (2002), 056306.
[8] F.F. Araujo, S. Grossmann, D. Lohse, Phys. Rev. Lett. 95 (2005), 084502.