Flow measurements in a model of the Czochralski crystal growth process

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An experimental study of the buoyancy-induced flow in a model of a Czochralski crystal growth system was conducted. Ultrasonic velocimetry was used to measure fluid velocities. To have similar thermal boundary conditions as in an industrial growth facility, a double walled glass crucible flown through by a heating fluid was chosen to hold the fluid. Similarity of the heat transfer conditions was achieved by selecting GaInSn as liquid metal under investigation. Due to the double-walled crucible, measurements through the container wall are difficult if ever possible. Since the availability of short ultrasonic transducers it is practicable to have the sensor immersed into the fluid. Measurements of the radial velocity component shortly below the melt surface across the entire diameter of the crucible at various azimuthal angles reveal the complex flow structure of natural convection in a Czochralski crucible. As it is not to be expected to grow high quality mono-crystalline crystals from a non-axisymmetric flow, rotating magnetic fields (RMF) are often proposed to render the flow more axisymmetric. This paper also addresses the question what happens to the buoyancy-driven flow when such an RMF is applied.

Keywords: Czochralski crystal growth, Rayleigh-Bénard convection, ultrasonic flow measurement, magnetohydrodynamics, electromagnetic stirring

1 INTRODUCTION

The mass production of mono-crystalline silicon by the Czochralski (Cz) technique has initiated a huge amount of research on the convection inside the crucible. Most of the investigations are numerical 2D simulations owing to the costly calculations in the full 3D case. Experimental work, on the other hand, is basically limited by two facts. Firstly, measurements in an industrial facility are hardly possible because of the high temperatures inside the crucible and restricted accessibility. Secondly, also physical modelling is expensive. This may be seen as the reason that a lot of work has been done on systems in which simplifications often might have been over-stressed in that they eliminated exactly those effects determining essential features of the flow in industrial installations.

One of such frequently made simplifications is that the flow in a Cz crucible is physically modelled by a Rayleigh-Bénard (RB) system. A cylindrical fluid volume, homogeneous upper and lower temperature boundary conditions, and an insulated side wall comprise this generic configuration. It is possibly not so much the differing geometry, a Cz crucible has a rounded bottom, that may cause another convective pattern than that to be found in an RB system, but rather the different thermal boundary conditions. A Cz system can, to a certain extent, be seen as being similar; one is concerned with natural convection due to a vertical temperature gradient ∇T_v. The crucible is heated from below, but also the side wall is heated - with the consequence that a primary horizontal temperature gradient ∇T_h exists in the same order as ∇T_v. Moreover, the variable heat flux through the quartz crucible wall into the melt differs substantially from the boundary conditions in the RB case. What holds for the lower boundary condition continues to be true for the upper one. In an RB system, the whole fluid surface is kept at a constant temperature, whereas the crystal in a growth facility covers only, between a quarter and a third of the diameter leaving the remaining majority of the surface open to the ambience.

Despite the lack of a deeper understanding of the flow in a Cz crucible stemming only from natural convection, means of flow control other than mechanical rotation have been subject of research for a long time - with the trend that this research is increasingly intensified. Amongst the numerous publications on RMF's being used to control the flow in a RB system [1-3] are referred to. The experimental study on the influence of an RMF in [4] was devoted to a modified RB system obeying the actual coverage of the surface by the crystal.

In its first part of the results, the present work attempts to fill the gap of missing fluid flow measurements in the mere buoyancy-driven case in a Cz crucible. Because the long-term perspective of the ongoing work is flow control by magnetic fields, the Cz model was mounted in a magnetic system providing, among others, also an RMF. The second part of the results is thus concerned with some preliminary results on the influence of electro-magnetic rotary stirring on the natural convection.

2 DESCRIPTION OF THE EXPERIMENT

The flow measurements were conducted in the Czochralski-like crucible depicted in Fig. 1. Heating was established by passing a temperature-controlled fluid from a thermostat through the spacing between the inner and outer vessel, which comprise the double-walled crucible. Extraction of heat through the growing crystal was simulated by a circular centrically mounted heat exchanger on top. Since the heat exchanger was made from copper, it imposes approximately isothermal boundary conditions in an area that corresponds to a fraction of coverage the same as in industrial installations. A sketch of the Cz setup including all relevant dimensions is also to be seen in Fig. 1. Adjustable by the filling level, the aspect ratio is determined by an effective height H_{eff} calculated from the fluid volume *V* and the area *A* in the majority of the upper part. It was a = 0.59 in the experiments.

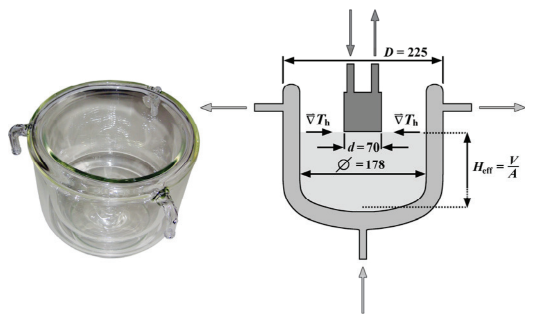

Figure 1: Photo and sketch of the Czochralski-like setup.

Flow measurements were done with a short ultrasonic transducer (8 MHz, type TR0805RS, Signal-Processing, Lausanne, Switzerland), a photo of which is provided in Fig. 2. A DOP2000 velocimeter from the same manufacturer was used to acquire the measuring data. For a description of the principle of operation of pulsed ultrasonic Doppler flow measurements, the pioneering work of Takeda is referred to [5, 6].

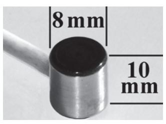

Figure 2: Photo of the short ultrasonic transducer that was immersed into the melt slightly below the surface to measure radial velocity sections.

Since it is hardly possible to measure through the double-wall of the crucible containing a liquid with different sound velocity and acoustic impedance, the transducer has to be immersed into the melt. In order to acquire as much of the radial profile as possible the transducer needs to be accordingly short in the measuring direction. It was then mounted on a x-y crossbar with an additional rotational unit so as to move it around the azimuth while the measuring direction was always slightly below the melt surface pointing towards the centre of the crucible.

To study the effect of an RMF on the natural convection in a Cz system, the whole apparatus was mounted inside the home-made MULTIpurpose MAGnetic field facility (MULTIMAG). This system offers, besides an RMF, linearly travelling, single-phase alternating and static magnetic fields, the latter in homogeneous or cusp configuration. MULTIMAG is described in detail in [7].

3 RESULTS AND DISCUSSION

The first natural question about the global flow structure in an axisymmetric experimental setup is whether the developing flow is also axisymmetric. Variation of the convective pattern in RB convection, i. e. in cylindrical containers with homogeneous upper and lower temperature boundary conditions, owing to the aspect ratio a was recently studied numerically and experimentally [8]. For $a \geq 0.63$ a single roll corresponding to an azimuthal wave number of $m = 1$ was found; fluid rising in a more or less kidney-shaped region at one side, moving along the surface to the opposing side, descending there, and closing along the bottom. This flow phenomenon often termed *wind* is frequently observed in RB systems.

As mentioned, the Cz system exhibits always significantly strong primary horizontal temperature gradients owing to the only partially cooled surface. The next question therefore is, if the instantaneously developing radially inwards directed flow due to these horizontal gradients, which is axisymmetric, will survive the *wind* when the vertical gradient overcomes the critical value for the onset of RB convection. The answer for a cylindrical geometry with $a = 1$, which aspect ratio is significantly above the border even of 0.63 found in [8], is given in [4]: it does not. For all temperature differences between the bottom heating plate and the top heat exchanger the *wind* came on top. It did not just occupy a center region while there was radial and axisymmetric inward flow in an annular region adjacent to the side wall - as in the generic RB configuration with full isothermal coverage of the surface, it penetrated the whole cylinder. The findings in [4], in conjunction with the fact that the crucible dimensions in an industrial Cz facility do not reach $a = 1$ even at the maximum initial filling level, was the motivation to conduct the present experiments with a about the threshold to mono-cellular convection. A fixed value of $a = 0.59$ was chosen (c. f. Fig. 1).

If it were possible to safely measure the whole radial section of velocity across the diameter, it would have been sufficient to move the transducer over 180° since measurements from opposing directions should yield the same result. Although quite a lot of profiles were measured and averaged for each azimuthal position, measuring from the opposing side at the same locations further diminishes statistical errors. More important, it is well known that the ultrasonic beam diverges while it moves away from the emitter; the farther the measuring location, the more the lateral size of the measuring volume increases over which the local velocity is averaged. So it is, at least, instructive to compare the results that have been obtained at, say, 0.5*R* from one side with those that were acquired at 1.5*R* from the other side.

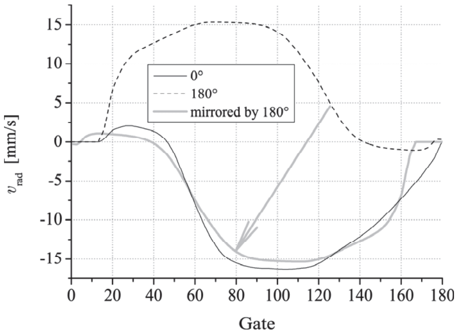

Figure 3: Radial velocity profiles across the whole diameter, measured from opposing sides with respect to the azimuthal angle.

Most important in the present case of the immersed sensor is that the sensor can not measure the location where it resides, continued by a further distance in beam direction where it does not work due to the ringing effect. This *dead zone* extents to about 15 mm, which is amply 8 % of the measuring depth. So, the information from the measurement done from the other side is vital. Fig. 3 contrasts two opposing velocity profiles acquired at 0° and 180°. The *dead zone* is obeyed by filling the missing gates with zeros.

Fig. 4 shows a series of radial velocity profiles with the azimuthal angle as parameter.

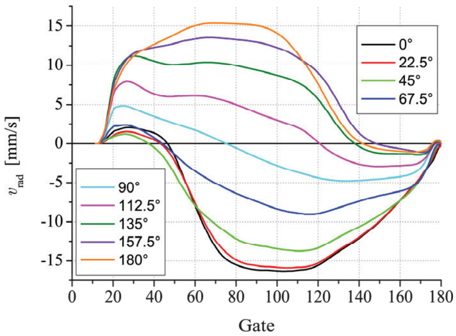

Figure 4: Radial velocity profiles across the diameter, measured in increments of 22.5° of the azimuthal angle.

If the flow in the Cz crucible were axisymmetric, all measurements at any azimuthal angle should look similar and something in between the measurements at 90° and 112.5°, crossing the $v = 0$ axis in the centre of the crucible. As this is not the case, the flow in the Cz crucible is certainly not axisymmetric, but rather three-dimensional on a global scale. What can be safely stated is, that the flow has a radially inwards directed component in an annular region adjacent to the crucible wall. Since this annular region does not penetrate the same distance from the wall towards the centre for any azimuthal angle, the observation in Fig. 4 may indicate a superposition of several azimuthal modes, one of which is the axisymmetric $m = 0$ one.

The top-view visualization in Fig. 5 attempts to give a pictorial impression of the convective pattern which is better suited to reveal insight into the complex flow structure in the Cz crucible.

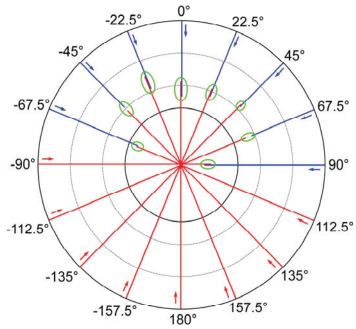

Figure 5: Top-view visualization of the flow structure in the Czochralski crucible. The details of how this plot was generated are described in the narrative.

Dropping the quantitative information from the velocities, a line is drawn from the crucible wall towards the centre for each measured azimuthal angle. The arrows indicate that the direction of flow is inwards there. The length of the lines is the distance from the wall until the velocity reverses sign. Blue is the colour for those lines, the distance up to the change of sign of velocity of which is less than the radius. In the case that the velocity changes sign in a distance longer than the radius, the line for that azimuthal angle is plotted in red. Consequently, each line from one side of the crucible wall to the opposing side of the crucible wall consists of two parts. The joint region of these two parts is encircled with an ellipse. Both profiles measured from opposing sides do not have the change of sign in velocity necessarily at precisely the same location due to some remaining uncertainties in the measurements. There can be a gap as in the case of the pair -45°/135°, or both parts of the profiles can overlap as in the case 0°/180°.

Since the ellipses are stagnation points of the radial velocity component and the flow is radially inwards directed away from the crucible wall everywhere, the region covered by these ellipses should fairly coincide with a downstream. Fig. 5 suggests that there is a mono-cellular $m = 1$ mode (wind) in the centre of the crucible superimposed to the axisymmetric mode described above. The mean direction of that *wind* is from in between -157.5° and 180° towards in between 0° and 22.5°. As the innermost black circle is the region of the top heat exchanger, the superimposed *wind* seemingly extends outside the crystal dummy towards the crucible walls, at least in its mean flow direction.

With the hindsight of knowledge about such a flow structure it becomes obvious why the crucible and the crystal in a growth facility are rotated; it should be difficult to grow a circular high quality single crystal from such an asymmetric flow. Since the temperature field is coupled to the flow field, means have to be sought which render the temperature distribution more axisymmetric. Instead of rotation of the crystal and the crucible another such means might be to rotate the melt by a rotating magnetic field, which is the next subject.

Describing the driving action of an RMF by the Taylor number [4, 7], two different RMF's were applied to the melt volume.

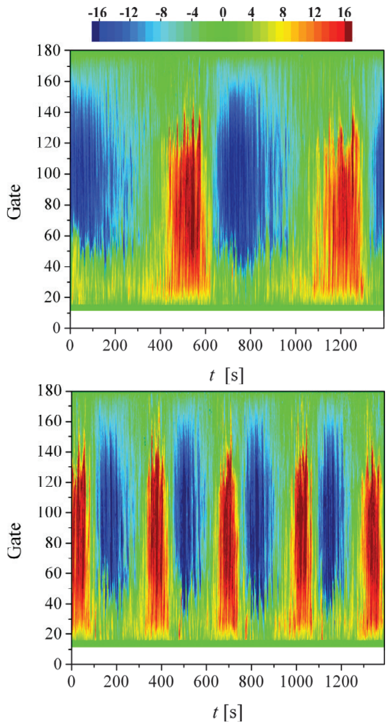

Figure 6: Time series of the radial velocity component measured at the azimuthal angle of 0°. The values of the legend of the velocities are mm/s. To the top Ta = $1.1{\cdot}10^6$ and Ta = $2{\cdot}10^6$ on the bottom.

In a first series, the Taylor number was kept at $1.1{\cdot}10^6$, a choice which was motivated by the parameters in [4]. In the modified RB system in [4], a cylinder with a partially cooled surface (crystal dummy), this Ta was in a range where the *wind* and rotary flow coexisted at all vertical temperature gradients. The top part in Fig. 6 shows that the *wind* rotates extremely slow at that Taylor number with about 670 seconds for one revolution of the fluid.

In the bottom part of Fig. 6, Ta was risen to $2{\cdot}10^6$. In [4] the mean temperature gradients were governed by the swirling flow evoked by the RMF. Here, the velocity time series show an increased rotational speed of the liquid metal, however, the strength of the *wind* is seemingly not affected by the swirling flow. The question about the correlation between the mean temperature gradients, which are an important parameter in crystal growth, and the flow field must remain open for the time being since temperatures have not been measured yet. Simultaneous flow and temperature measurements will be subject of ongoing work.

4 SUMMARY

Subject of investigation was the natural convection in a Czochralski crystal growth model, for which reliable velocity data have not been available to the best of our knowledge. Velocity profiles acquired across the melt surface at various azimuthal angles indicate that the convective pattern is a superposition of an axisymmetric m = 0 mode and a mono-cellular m = 1 mode. The latter, often called *wind*, is restricted to the central region of the crucible.

When applying a rotating magnetic field, the *wind* rotates slowly. The revolution rate increases with the strength of the magnetic field, however, it does not lose vigour at field strength where the temperature distribution is strongly affected by the swirling flow. Future work will thus address two issues: (i) Simultaneous temperature measurements are needed to shed some light onto the correlation between temperature gradients in the growth zone and the flow. (ii) Increased field strengths have to be investigated to answer the question whether it is possible to suppress the buoyant convection by rotary stirring completely, that is to say whether it is possible to render the flow axisymmetric.

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