MODEL EXPERIMENTS FOR INVESTIGATIONS OF HEAT TRANSFER PHENOMENA IN THE CZOCHRALSKI CRYSTAL GROWTH

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Abstract: A low temperature liquid metal model of the Czochralski crystal growth process is considered experimentally under conditions of high aspect ratio. We focus on the influence of a rotating magnetic field (RMF) and/or crystal rotation on temperature fluctuation near the crystal edge. The radial flow structure is observed by ultrasound Doppler velocimetry (UDV). It is concluded that the effect of RMF on the temperature fluctuation is less expressed than in a Rayleigh-Bénard cell.

1. 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. The fluid flow inside the crucible serves as a carrier for mass and temperature and affects crucially the quality of the growing crystal.

The RMF is known to efficiently suppress the Rayleigh-Bénard (RB) instability in a liquid metal cylinder of a size that is relevant for industrial Cz growth [1]. It has been speculated that this RMF property may better stabilize the melt than the Cz crucible rotation allowing, thus, to increase the initial filling height. The current study aims to test this hypothesis in a low temperature model of the Cz process. This is why we use a melt aspect ratio $a=H/D=0.59$ that is much higher than usual in industry. Besides, this value marks the transition between axisymmetric and mono-cellular convection in the RB cell [2].

The existence of a triple point in the Cz system is one of the most important differences to the RB cell. This is the point where the free surface of melt meets the growing crystal. Flow and temperature conditions at this point have a particularly profound effect on the quality of the end product. Therefore, our study is focused on measurements in the vicinity of this point. For details concerning the relevant theoretical background and the experimental setup we refer to [2,3].

2. Experimental setup

The central part of the setup comprises the double-walled glass crucible (diameter D) filled up to a height H with liquid GaInSn, where H is equal to the melt volume divided by the top surface area. A photograph of the crucible is shown on the left side of Fig. 1. Heating was controlled by passing thermostated silicone oil through the wall gap. The growing crystal is simulated by a co-axially mounted copper cylinder cooled by thermostated water (so-called cold finger). The cold finger is immersed about 3 mm into the liquid GaInSn.

Two versions of temperature boundary conditions are realized at the free melt surface: (1) air-cooled surface with practically negligible heat loss, and (2) water cooled surface as shown in Fig. 1. The second version is meant to better model radiative heat loss in the Cz process. For purpose of surface protection from oxidation a weak aqueous solution of KOH is added to the surface of GaInSn. This layer is cooled by thermostated copper coils.

As the experiment was demounted in the interval between the measuring campaigns conducted for both versions of surface cooling, additional measurements were carried out to check the reproducibility of the results (see next section). For applying an RMF for electromagnetic flow control in the crucible, the experiment was mounted inside the coil system MULTIMAG, whose features are described in [4].

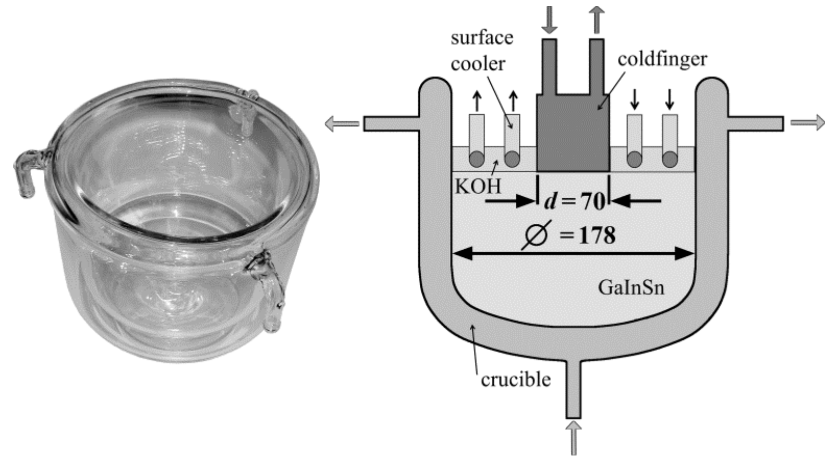

Figure 1: The left photo depicts the double-walled glass crucible. The right part shows a sketch of the experimental setup with the cold finger modelling the growing crystal and the additional surface cooler.

3. Results

The UDV technique was used to measure the radial component of the fluid velocity slightly below the free surface. The ultrasonic sensor was immersed into the melt at several azimuthal positions close to the inner rim of the crucible. The first series of experiments was performed in a setup version with the air-cooled free surface. The flow measurements were repeated several times in order to check the reproducibility of the results. Fig. 2 displays the radial component of the fluid velocity measured beneath the free surface. The grey circle marks the area which is covered by the cold finger. The measurements reveal a complex flow structure which may occur similarly in a Cz crucible. A converging flow can be observed at the surface indicated by the arrows at the inner rim of the crucible. However, the flow pattern is not axisymmetric. The thick black lines illustrate a dominating flow direction which extends beyond the axis whereas the thick grey lines show an inwards flow which does not reach the axis of the crucible. The ellipses highlight the zones where a conversion of the flow direction was observed.

Fig. 2 compares two different flow regimes measured before and after rebuilding the experimental facility, respectively. A stable flow structure, Fig. 2a, was observed during the first measuring campaign [2]. The flow measurements shown in Fig. 2b were performed after the reconstruction. The additional surface cooling was not yet installed in this case, so that identical thermal boundary conditions can be assumed. On the contrary, the flow regime shown in Fig. 2b developed occasionally, remained for several hours and changed then again to the first one. The reason for such random changes between these two regimes is not yet clear. Although big efforts were made to reproduce identically the first setup, remaining uncertainties such as axis tilting or slightly different filling levels of the GaInSn may cause these effects. However, both flow regimes show a comparable behaviour, the major difference is a rotation of about 160° around the centre axis.

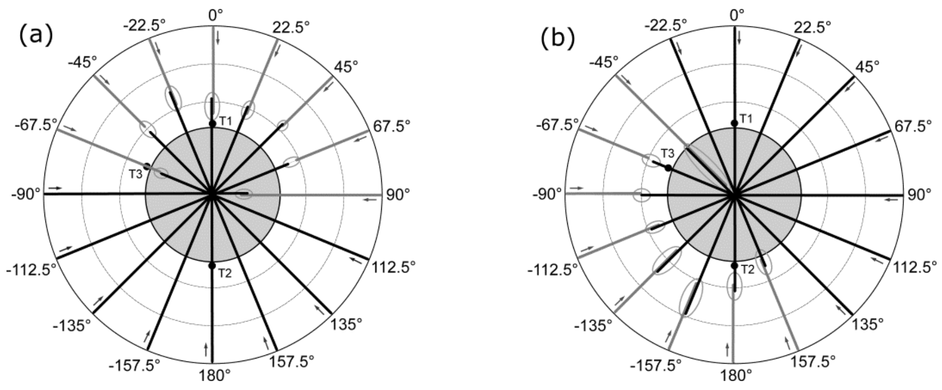

Figure 2: Top-view visualization of two flow regimes just below the surface. (a) as measured in the first setup (cf. Fig. 7 in [3]); (b) second setup but still without the surface cooler. T1 to T3 indicate the places where thermocouples were positioned.

The following temperature measurements reported here were carried out with the activated surface cooler. Motivated by the UDV measurements, three distinct azimuthal positions (cf. positions T1-T3 in Fig. 2.) were chosen to monitor the temperature near the triple point. Temperature time series recorded in the pure buoyant case (without RMF or rotation of the cold finger) are shown in Fig. 3. The differences with respect to the absolute value of the temperature and the amplitude of the temperature fluctuations demonstrate the non-axisymmetric character of the flow.

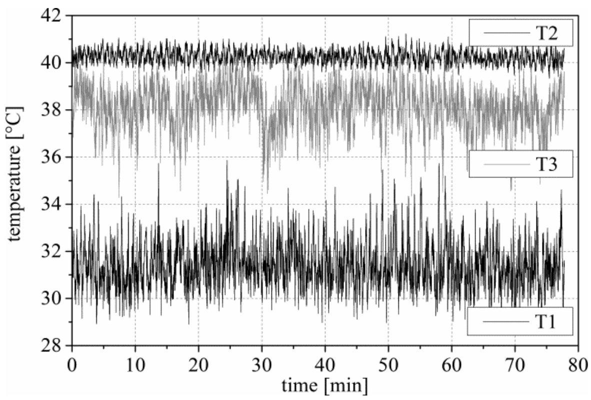

Figure 3: Temperature time series in the pure buoyant case showing the non-axisymmetric character of the flow. T1, T2 and T3 indicate at which positions the signals were recorded.

The temperature fluctuations detected at the positions T1 and T3 are much more pronounced compared to the signal recorded at position T2 and indicate, thus, the flow regime shown in Fig. 2a may be present in this case. The measuring positions T1 and T3 are located close to the zone where opposite streams of the radially inward flow collide. This zone is characterized by a highly turbulent flow. In contrast, the ascending fluid at the azimuthal position 180° flows towards the centre, reaches position T2 and passes the cold finger without distinct perturbation. Therefore, the fluctuations in the temperature are there less compared to the other positions. Under the cold finger the fluid cools down and meets, after passing T1, the fluid coming from the opposite side at the azimuthal position 0°.

From the knowledge about such a flow structure it becomes comprehensible why the crucible and/or the crystal in a growth facility need to be rotated. It would be difficult to grow a circular, high quality single crystal under such asymmetric flow conditions. In order to approach a more axisymmetric temperature distribution, the rotation of the crystal is applied as a method to modify the temperature field. An alternative method in comparison to mechanical rotation can be the generation of a melt rotation by applying an RMF. The impact of an RMF is described by the dimensionless Taylor number Ta [2, 3].

Whatever kind of rotation is applied to the system, the thermal structures will be transported by the fluid flow. Fig. 4 shows temperature measurements for both cases of cold finger and RMF rotation. The time series were recorded at the position T2. Furthermore, the rotational parameters applied were varied in a wide range: Ta between 2.0×10^5 and 2.0×10^8; the revolution rate of the cold finger from 5 up to 55 rpm. In both cases a characteristic oscillation frequency of the temperature signal was observed which indicate the advection of the thermal structures. The diagram on the left side in Fig. 5 summarizes these results.

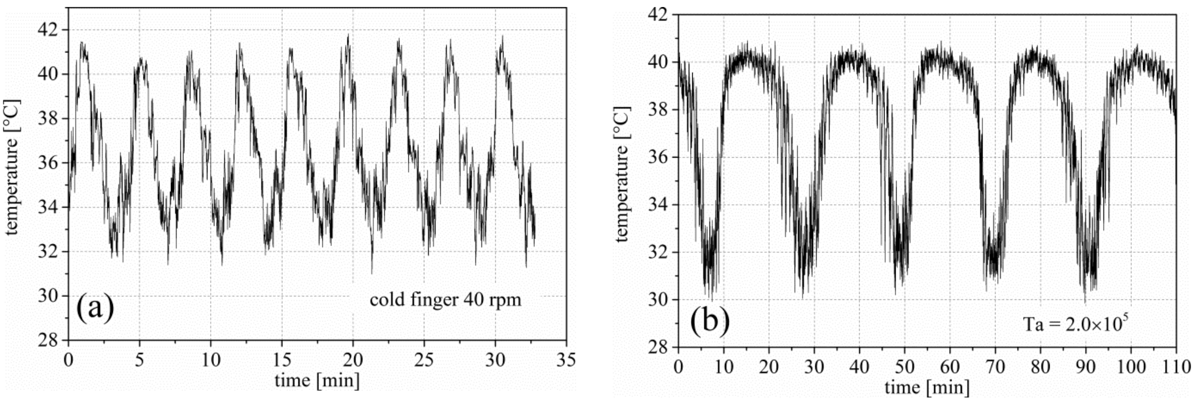

Figure 4: Measured temperature time series at T2 under the influence of (a) cold finger rotation, and (b) of the RMF. Similar large oscillating temperature structures occur at T1 and T3 too, but with a certain phase difference caused by the rotation of the fluid.

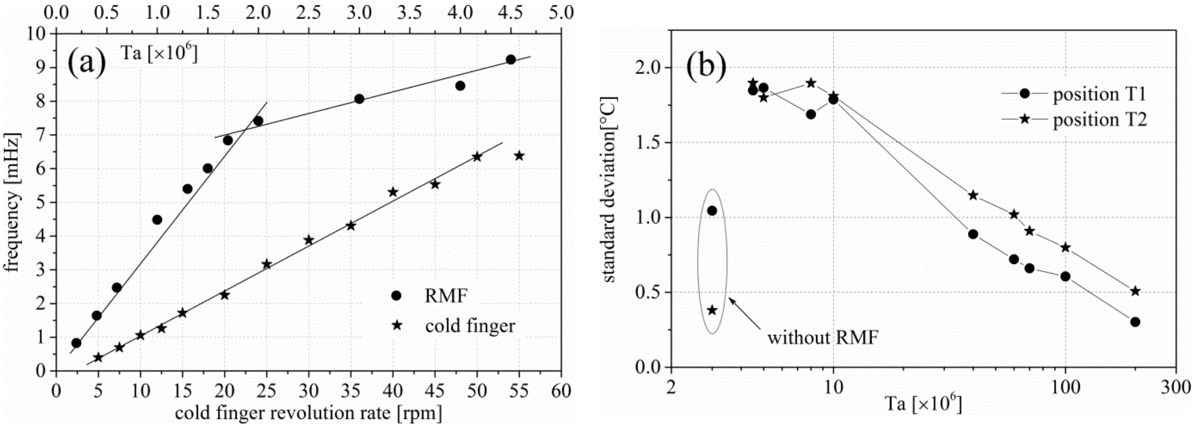

Figure 5: (a) Rotation rate of the thermal structures as function of the cold finger revolution rate (lower abscissa) and of the Ta number (upper abscissa). (b) Standard deviation of the temperature fluctuations in the higher Ta number range after break down of the regular thermal structures.

The oscillation of the thermal structures was observed up to the maximal possible cold finger revolution rate. In contrast, for application of an RMF at Ta numbers up to approximately 4.5×10^6 two “scaling” ranges were detected. Going to higher Ta numbers beyond 4.5×10^6 the previously observed regular thermal structures break down. With increasing Ta, cf. diagram in Fig. 5b, the standard deviation of the temperature fluctuations decrease and reach typical values found in the mere buoyancy case for relatively high Ta numbers.

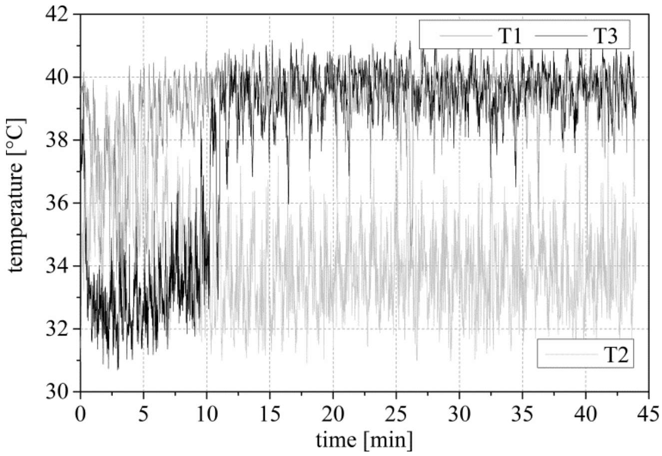

Figure 6: Temperature time diagram for a combination between an RMF and an oppositely rotating cold finger.

It is clear that thermal structures such as shown in Fig. 4 may not be very beneficial for the growth of high quality single crystals. However, a combination of the RMF forced rotation and an oppositely rotating cold finger appears to be a promising approach to achieve a symmetric flow without distinct large-scale temperature fluctuations. Such a measurement is shown in Fig. 6. In this situation, an RMF with $Ta = 6.0\times10^5$ (equivalent to a thermal structure rotation rate of 2.5 mHz as seen in Fig. 5a) was first applied to the melt. Then, the corresponding revolution rate of the cold finger with 21 rpm was switched on (t = 0min). Fig. 6 demonstrates that the large thermal structures disappear after approximately 10 to 15 minutes.

4. Summary and outlook

Flow velocities and temperatures are measured in a Cz model with a high aspect ratio. Influence of crystal and/or RMF driven melt rotation on temperature fluctuation is investigated in vicinity of the triple point. Separately applied RMF stirring or crystal rotation causes large scale rotating structure that is an inferior condition in crystal growth. An appropriate combination of the RMF and crystal rotation brings this structure to standstill. It is concluded that the stabilizing RMF effect in the present Cz model is much less expressed than in an RB cell.

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5. References

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