Experimental investigation on the buoyancy-induced flow in a model of the Czochralski crystal growth process

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Abstract
Within this paper we present a model experiment focusing on investigations of the flow field in a Czochralski puller. Low melting point liquid metals as GaInSn are an important tool to investigate the flow structure for such industrial processes. The topology of the prevailing thermally-driven convection might be very complex and is mainly determined by the aspect ratio of the liquid volume and the strength of the convection described by the characteristic dimensionless Grashof number. The measurements of the fluid flow have been conducted by means of the ultrasound Doppler velocimetry (UDV) with and without the influence of external magnetic fields. Two kinds of sensor configurations were used to investigate the flow. Firstly, measurements of the radial velocity component by means of single UDV transducers were carried out shortly below the melt surface across the entire diameter of the cylindrical liquid column at various azimuthal angles. Secondly, a vertically arranged UDV array was applied at the side of the cylinder to obtain more detailed information about the radial velocities in the covered meridional plane. The results reveal the complex flow structure of natural convection in a Czochralski crucible which gains in complexity with applied external magnetic fields.

Keywords: Czochralski crystal growth, Electromagnetic stirring, Rayleigh-Bénard convection, Flow measurements

Introduction
Mono-crystalline silicon crystals are, nowadays, required in many electronic devices and for the power generation by photovoltaic systems. One of the main objectives in the manufacturing process of photovoltaic silicon wafers by the Czochralski (Cz) method is to produce crystals with low concentration of impurities such as oxygen, carbon and/or other materials. These adverse impurities might stem from the crucible and/or other inner parts of the crystal pulling equipment, transported by the melt flow and incorporated in the solidification process. Thus, the quality of the growing crystal and the yield of the wafers are crucially affected by the fluid flow. Consequently, the mass production of mono-crystalline silicon by the Cz technique has initiated a great effort of research on the convection inside the crucible. But reliable flow measurements at the real process are practically nonexistent and the validation of numerical models can only be accomplished by laboratory model experiments. Moreover, further optimizations of the process by means of applied magnetic fields are mainly based on numerical simulations of the flow, heat and mass transfer, and electromagnetic fields. Thus, the experimental modeling of the process on a laboratory scale still remains a valuable tool for the near future. For details on the growth of semiconductor silicon crystals the reader is referred to [1].

Modelling of the Czochralski technique
The simplest model of the Cz facility considers a Rayleigh-Bénard (RB) configuration, in particular a cylindrical cell characterized by a height $H$ and diameter $2R$ with adiabatic insulated side walls. The cell is heated from the bottom and cooled at the top. The temperature gradient $\Delta T$ between the bottom and top sides generates buoyant convection. The dynamics of the mere thermally induced convection may be described by mainly three control parameters. The dimensionless Rayleigh number $Ra$ is the crucial parameter in modelling buoyancy and describes its strength. Instead of $Ra$ the Grashof number $Gr$ is also commonly used for this purpose and is coupled to $Ra$ by the second control parameter, the Prandtl number $Pr$, according to $Ra = Gr \times Pr$. Generally, molten metals and semiconductor melts are low $Pr$ number fluids in the order of $10^{-2}$, which means that the thermal boundary layer thickness exceeds the viscous boundary layer by a factor in the order of 10. 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 [2,3]. The initial filling level in the real industrial Cz facility does not reach $a = 1$, it is even lower than $a = 0.5$ and decreases continuously during the process.

Other control parameter may occur if external magnetic fields are exposed to the system, such as the Taylor number $Ta$ in case of the rotating magnetic field (RMF). The relevant parameter, $Ta$, describes the driving effect on the melt under investigation, which is primarily of swirling type but also causing a meridional secondary flow [4,5].
Experimental setup

The present experimental setup involves a modified RB configuration, a cylindrical melt column of variable aspect ratio homogeneously heated from the bottom. The left photo in Fig. 1 illustrates the experimental setup mounted inside the magnetic coil system MULTIMAG [6]. Besides the already mentioned RMF, it offers a variety of other magnetic field types, such as the traveling (TMF), pulsed (PMF), vertical DC or vertical DC of cusp type, and even linear superposition of them. As working fluid the ternary alloy GaInSn [7] was used because it remains liquid at room temperature and as distinguished from mercury it is non-poisonous. Moreover, its low Pr number is similar to that of molten silicon.

The heater 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 to monitor its temperature. The upper thermal boundary condition in a Cz system is realized by a partially cooled surface. The partial cooling in our experiment covers approximately the same fraction area as the crystal does in an industrial Cz 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 rotatable. The precise temperature control of the finger is guaranteed by a coolant provided from a thermostat with a large reservoir and high flow rate. Moreover, the temperature of the cold finger is monitored at various positions. During the measurements, the apparatus was embedded in mineral wool to minimize the lateral heat loss.

Flow field measurements were performed by the UDV technique. Its operation principle is described in the pioneering work by Takeda [8]. Mainly two features approve the application of UDV indispensable for the present work. Firstly, it works in opaque media including liquid metals. Secondly, it allows the quasi-simultaneous measurement of an entire profile of the local velocity component in direction of the sound propagation along the ultrasonic beam. Two different sensor arrangements were used. Most of the measurements were taken by a DOP2000 velocimeter (Signal-Processing, Lausanne, Switzerland) with single ultrasound transducers. As it can be seen on the middle part of Fig. 1 six single sensors were arranged at different azimuthal positions, each one differing by 30° to the next one. This allows gathering flow information over the whole circumference of the vessel. Furthermore, the different mounting levels allow measurements for variable aspect ratios. On the right in Fig. 1 a second setup with an attached transducer array can be seen. The UDV array consists of 25 single transducer elements arranged linearly, each of a size of 2.3×5 mm² with an element pitch of 2.7 mm. Thus, it allows flow measurements of the velocity component perpendicular to the transducer surface over a field width of 67 mm. Moreover, multiple array arrangements would allow measurements of further velocity components and, therefore, enable complex flow mapping in the covered plane [9,10].

Results

For the present study flow velocity measurements were performed at mainly two different aspect ratios, \(a = 0.45\) (\(H = 80\) mm) and \(a = 0.3\) (\(H = 54\) mm), and under the influence of different types of magnetic fields (RMF, TMF, cusp). Nevertheless, only a small cutout of some selected aspects can be presented in this paper.

In the range \(1.4\times10^4 \leq Gr \leq 1.6\times10^4\) two different flow regimes were identified and investigated for \(a = 0.3\). The vertical position of the sensors was thereby at \(h = 40\) mm. For a low temperature gradient of \(\Delta T = 1\) K (\(Gr = 1.6\times10^4\)) an axisymmetric flow structure occurs. The fluid heated from the bottom rises along the rim, continues towards the center axis, drops in the central region below the cold finger and closes outward to the rim. Fig. 2 on the left illustrates such a measurement done with one single transducer at the randomly selected azimuthal position \(\varphi = 0°\). Already for slightly higher \(\Delta T = 3.1\) K (\(Gr = 5.1\times10^4\)) the axisymmetric flow topology becomes unstable (cf. Fig. 2-right) and for further increased \(Gr\) (\(Gr = 4.2\times10^4, \Delta T = 22\) K) the flow develops the wind structure which is a large scale single roll circulating inside the vessel. Such a structure is well-known in general RB systems and frequently observed for \(a = 1.0\).

It should be mentioned here that the preferred main flow direction of the wind develops randomly and is influenced mainly from imperfections of the experimental setup. Fig. 3 shows two time series measurements in the wind-mode at the positions \(\varphi = 0°\) and \(\varphi = 90°\). The latter one corresponds thereby to the main circulation loop of the flow.
Fig. 2: Time series and the related mean profile of the radial velocity component in the axisymmetric case (left) and in the transitional range (right).

Fig. 3: Time series observed for long term measurements in the wind-mode.

Fig. 4: Mean radial velocity profiles across the diameter for the wind-mode (left) and typical surface integrals of the mean profiles for different measurements (right).

The orientation of the wind becomes clearer from Fig. 4. The left diagram shows the mean radial velocity profiles at each of the azimuthal positions and the right side the surface integral of the respective profile. The maximum in the curve at approximately $\phi = 90^\circ$ indicates clearly the main circulation loop of the wind. Furthermore, the right part of Fig. 4 summarizes three different measurements performed at various days and demonstrates well the stability of the wind. It must be mentioned, however, that this was not the case for $a = 0.45$. Although not shown here, the wind was unstable in the sense that the maximum of the integral curves varied in time over a large azimuthal range, even flow reversals were occasionally observed.

Fig. 5: Radial flow maps measured using the UDV array. Arrows indicate the radial flow direction and are not scaled. Due to design related reasons the lower 20 mm could not be measured.

Fig. 6: Influence of the RMF on the wind-mode measured at $\phi = 0^\circ$ for two different, relatively low strengths of the RMF (upper part). The lower diagram shows the case for a strong RMF.

As previously mentioned, some selected measurements were done by means of the UDV array technique in a second setup. Due to the ability to measure quasi simultaneously along several sensor lines, a more informative radial flow map is achieved. Fig. 5 shows in the upper diagram such a purely buoyancy driven radial flow map in the axisymmetric low $Gr$ case and for the fully developed wind-mode in the lower part. Furthermore, from the lower diagram it is apparent that the axial-radial measurement plane of the UDV array does not fully correspond with the main circulation loop of the flow. Application of an RMF to the wind-mode causes a co-rotation of the wind (cf. upper part of Fig. 6). For a
sufficiently high value of $Ta$ the wind completely disappears as it can be seen in the lower diagram of Fig. 6.

Fig. 7: Radial flow under the influence of different RMF in the axisymmetric case measured in the single transducer configuration at $h = 40$ mm.

Fig. 8: Mean radial flow maps under the influence of two different RMF’s.

A different influence of the RMF is observed by applying it to the axisymmetric case. The diagrams in Fig. 7 visualize the single sensor measurements for three different $Ta$ numbers. At first glance, common to the diagrams are the first few minutes in which the RMF was not yet switched on. The axisymmetric structure is clearly observed here. When the RMF is switched on, some minor differences are observed between $Ta = 1.0 \times 10^6$ and $Ta = 1.4 \times 10^6$. For $Ta = 2.6 \times 10^6$ the axial symmetry disappears, rather a similarity to the wind structure is observed – at least along the single sensor measurement line ($h = 40$ mm). Of course it might be somewhat misleading to conclude from a single line measurement to the overall flow structure. In this regard, the major feature of the UDV array to measure simultaneously several lines becomes rather advantageous. The diagrams in Fig. 8 illustrate the mean radial flow maps for the same $Gr$ and $Ta$ numbers as in Fig. 7. By increasing the RMF strength rather a kind of squeezing occurs and the inward flow at the upper part of the vessel concentrates more and more to the region below the surface. Since the diagrams in Fig. 8 are the result of an averaging process over the measuring time of about 1 hour, the flow still has a time dependent and complex behaviour. Thus, care has to be taken by interpreting the overall flow topology.

Summary
A Cz-like crystal growth model exposed to different magnetic fields was the object of the present investigation. Ultrasound measurements of the fluid flow were performed by varying the strength of the RMF ($Ta$) and that of the buoyancy ($Gr$). Besides the single ultrasound transducers, the UDV array might provide a deeper insight into the complex flow topology in a Cz setup. Furthermore, the experimental data here serve as a benchmark object for numerical codes which are still under development.

Acknowledgment
Financial support by the German Federal Ministry for Economic Affairs and Energy in the framework of the KORONA project is gratefully acknowledged.

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