Experimental investigations on thermal, thermocapillary and forced convection in Czochralski crystal growth configuration

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Abstract

One of the greatest technological and scientific challenges of crystal growth is to achieve homogeneity of the material properties (e.g. electrical resistance in the micro range). According to the present know-how, the cause of fluctuations of the electrical resistance is due to fluctuations of the growth rate of the crystal. One of the possible causes for fluctuations of the growth rate is the instationary convection in the melt. The focus of the project is to examine the influence of the thermal Marangoni effect on the temperature and velocity field due to spatial and temporal variations of the temperature field adjusted from the outside. Moreover, the aims of the project are to analyse the stabilisation effect of rotation on thermocapillary flow, to determine the start of oscillatory motions and to examine the transition of an oscillatory mode to higher modes as well as the transition to turbulent motion.

1 Introduction

The application of monocrystals enormously increased during the recent decades. The quality of the monocrystals mainly depends on the “interface and transport phenomenon” occurring in the melt during the production process. One of the possible causes for fluctuations of the growth rate is the instationary convection in the melt, generated by the thermal and kinematic boundary conditions. The investigation of the convection under micro-gravity conditions, for example in a fall tower, during parable flights or in space laboratories, is of great importance because the boundary convection can be studied independently of the natural convection. The project is based on investigations of boundary driven convection in a Czochralski configuration, which is presently used to produce about 80% of the silicon monocrystals world-wide. The aim of the experiments is to investigate the interaction of the thermal, thermocapillary and forced convection in a modified Czochralski configuration in order to detect new flow configurations which may help to find possibilities for the systematic control of the convection and in this way to tray and make suggestions of how to optimise the production process.

2 State of the art

Nowadays there are ways to numerically calculate the stationary boundary convection in advance knowing the boundary conditions and using commercial software. Making theoretical predictions is almost impossible if oscillatory instabilities or turbulent transition are considered, because the understanding of these phenomena is not inefficient yet.

Due to the complexity of the problem, point measurements are not sufficient for understanding the flow pattern and for comparing experimental data with numerical simulations. Hence, full field measurements of velocity and temperature gained great importance for the quantitative evaluation of thermally driven flows. To achieve this objective, a new experimental technique, Particle Image Thermometry (PIT) based on an analysis of the colour and displacement of thermochromic liquid crystal tracers (TLC) was applied to determine both the temperature and velocity fields of the flow (Kowalewski et al [2], Szymczyk et al. [4]). It combines PIT and Particle Image Velocimetry (PIV). Full 2-D temperature and velocity fields are determined from a pair or a longer sequence of colour images taken for the selected cross-section of the flow.

2.1 Theoretical and experimental activities

Mihelcic, Wenzl and Wingerath [3] published several papers about the Czochralski method. They confirmed the statement of Szymczyk [4] that the instability on the fluid/gas surface occurs immediately after the radial temperature gradient has been created on the interface of the melt. In addition the instability greatly depends on the geometrical parameters. In an other paper Szymczyk [5] confirmed the well-known fluid mechanics behaviour, that fluids with very low Prandtl–Numbers show strong tendency for transition into oscillatory or turbulent flow conditions. Japanese scientists [6] verified that the Péclet–Number, the dimension- free velocity of the crystal growth, with non-oscillating surfaces is much higher than with oscillating and that the deviation between different conditions has a large influence on the length and the diameter of the crystal. They also developed a software for the three-dimensional calculation of the thermal tension in the Czochralski crystal-growth process of GSO (Gd_2SiO_5) [7].

2.2 Experimental investigations on crystal-growing equipment

The joint numerical and experimental investigations of the Universities of Madrid and Minnesota [8], [9] have proved that the temperature gradient has the largest influence on the morphology of the crystal's exterior surface. Lee and Chun [10] from the Pohang University of South Korea investigated the waves on the melt surface during rotating Czochralski crystal-growth, which appears by means of magnetic fields induction.

3 The experimental equipment

The experimental equipment was designed in a way, that it best matches the real Czochralski-process (Figure 1). To provide the thermal and kinematic boundary conditions, the experimental equipment allows setting the temperature of the stick, surrounding fluid bath and measuring cubicle bottom. There is also the possibility of heating the upper margin of the experimental fluid, which is considered to be a modification of the real process. In addition, it is possible to rotate the measuring cubicle and the stick independently of each other at variable speed and direction of rotation. Different glycerine-water mixtures are used as experimental fluids.

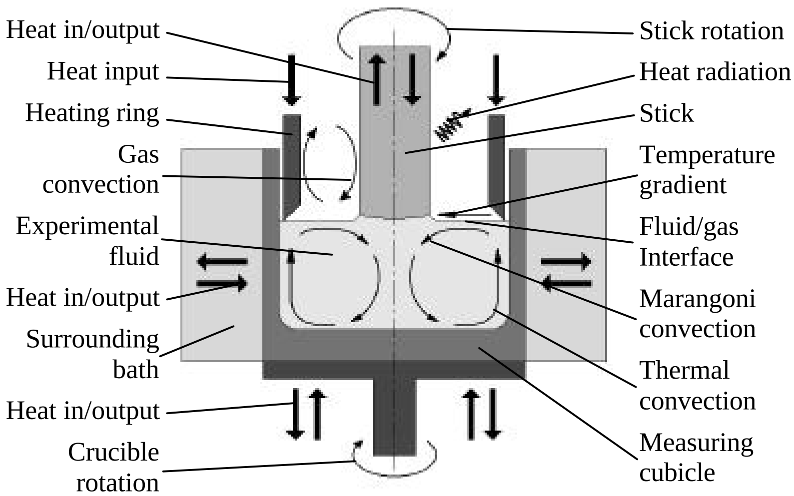

Figure 1: Schema of the modified Czochralski configuration

4 Optical flow measurement systems

Two optical measurement systems are simultaneously used to visualise the flow process (Figure 2). Using the Particle Image Thermometry in combination with liquid crystals the temperature field has been determined qualitatively and quantitatively. Using the Particle Image Velocimetry in combination with liquid crystals, which are here used as tracers, the velocity field has been measured quantitatively. The fluids used in the experiments behave similar to the real melt from a fluid mechanics point of view. Thus the results of the thermal and kinematic influences of these experiments allow conclusions regarding the realisation of the technological process to be drawn .

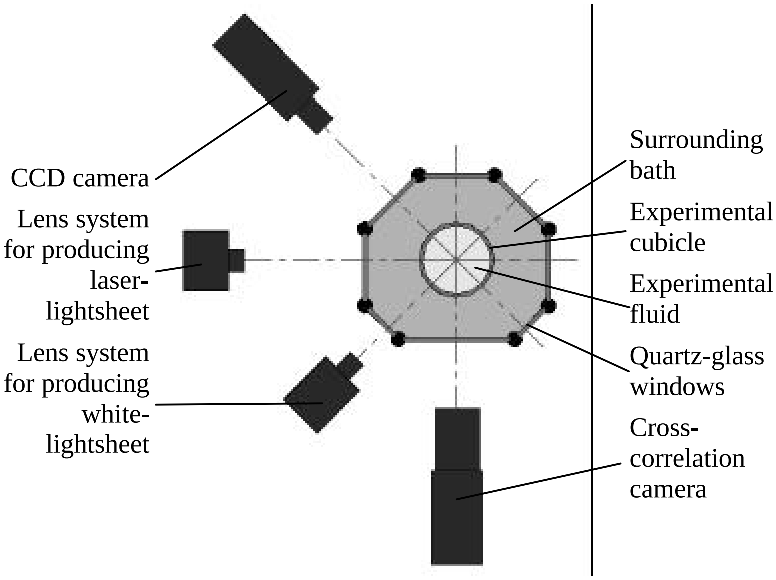

Figure 2: Lightsheet systems layout of the experimental equipment

4.1 Particle Image Thermometry (PIT)

The Particle Image Thermometry is based on the property of certain liquid crystals to reflect light in different colours depending on their temperature and the observation angles. The temperature field is visualised using a halogen or xenon lamp and the images are recorded with a CCD camera. The liquid crystals are characterised by their size, start and end temperature and their density. The start temperature, so called Red Start (RS), is the temperature of which the crystals begin to reflect red light. As the temperature increases, the crystals reflect the whole visible band of the white light colour spectrum beginning with red, over yellow, green and blue to violet. When reaching the end of the spectrum, i.e. the end temperature or Colourless (CL), the fluid becomes colourless.

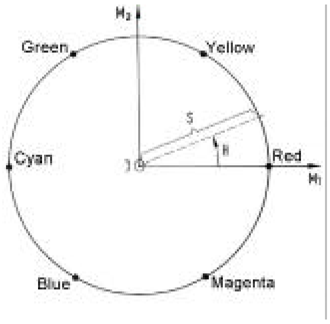

Figure 3: Graphical presentation of the HSI colour model (Haberbäcker [11])

The temperature measurement is based on the Red-Green-Blue (RGB) images recorded with the camera. Only knowing this RGB information, there is no explicit correlation between colour and temperature possible. To establish this correlation the Hue-Saturation-Intensity (HSI) model for temperature measurement should be used. The HSI model is based on twisting the RGB coordinate system and the presentation in cylindrical coordinates. The Hue (coloration) is the pure light colour, i.e. the dominating light colour, which can be seen. It represents the dominant wave length which is directly dependent on the temperature (Figure 3). Thus an explicit correlation between temperature and colour can be established. The Hue is specified as an angle, which in practice reside in the interval [0,255]. In this way the colour red is assigned the Hue value of 0, the colour yellow of 42.5 and the colour blue of 170. The saturation is a parameter for the dilution of a colour with white. The intensity is a colour-neutral attribute and describes the relative brightness. The conversion from the RGB to the HSI system can be made using the following formulas:

$$Hue = \frac{1}{2p}\arctan\left(\frac{2R - G - B}{\sqrt{3}\cdot(G - B)}\right)$$

$$Saturation = \sqrt{R^2 + G^2 + B^2 - RG - RB - GB}$$

$$Intensity = \frac{1}{3}(R + G + B)$$

The dependence of the Hue on the temperature is strong by non-linear and is described with a so called calibration curve (Figure 4). The calibration curve is dependent on the above mentioned factors, like experimental fluid, viewing angle, the type of the used light or light source. When changing one of these factors, a new calibration has to be done and a new calibration curveis obtained. The images are recorded, the temperature is measured and the associated Hue is assigned. It is very important to achieve an exact and correct correlation between the temperature and the Hue. The more sampling points (temperature/Hue) are acquired, the more exact calibration curve will be. The calibration polynomial has the form:

$$t(h) = a_n \cdot h^n + a_{n-1} \cdot n^{n-1} + ... + a_1 \cdot h^1 + a_0$$

t(h) is the temperature as a function of the Hue and a_n are the polynomial coefficients of the calibration polynomial. They are calculated by the FanHue program developed by us. By drawing the polynomial trough a number of sampling points, it may happen that it oscillates between some of them. To verify this, the calibration polynomial has to be graphically presented in any case. This can be done using FanHue or MathCad. If there are any oscillations, the degree of the polynomial has to be reduced.

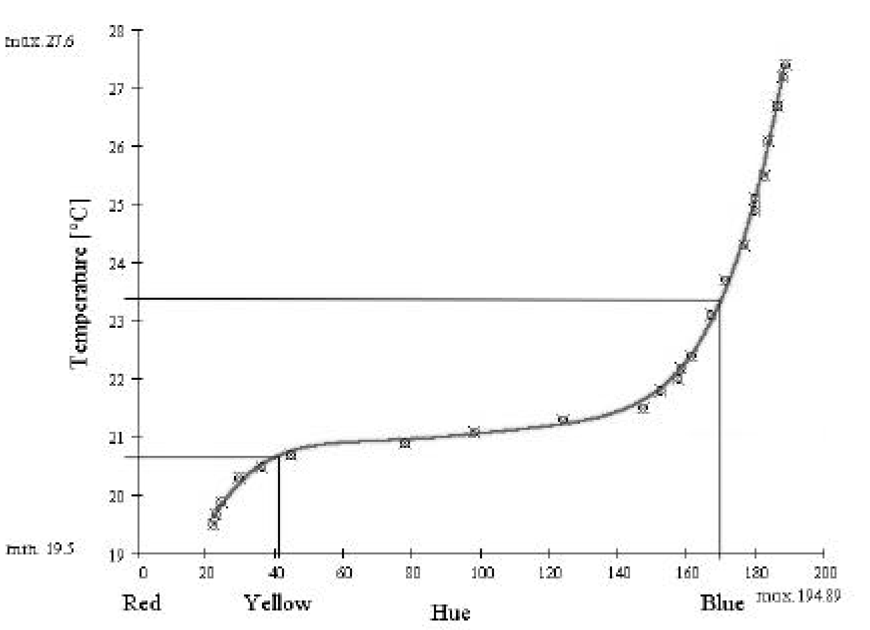

Figure 4: Calibration curve for KXN 2030 liquid crystals

The analysis of the temperature field images begins with a conversion into the BMP format. Then the images are processed using the FanHue program. The ultimate isotherms are illustrated using TecPlot. The images analysed by FanHue are processed pixel-wise and are not “blurred” or filtered, so the first plots made by TecPlot are not much meaningful. The isotherms are easily readable only after smoothing them.

4.2 Particle Image Velocimetry (PIV)

The Particle Image Velocimetry allows contactless measurement of the flow velocity of the whole field. After generating a lightsheet, which binds the measurement volume, it is possible to visualise the flow process and in this way to calculate the flow velocity.

5 Presentation of the results

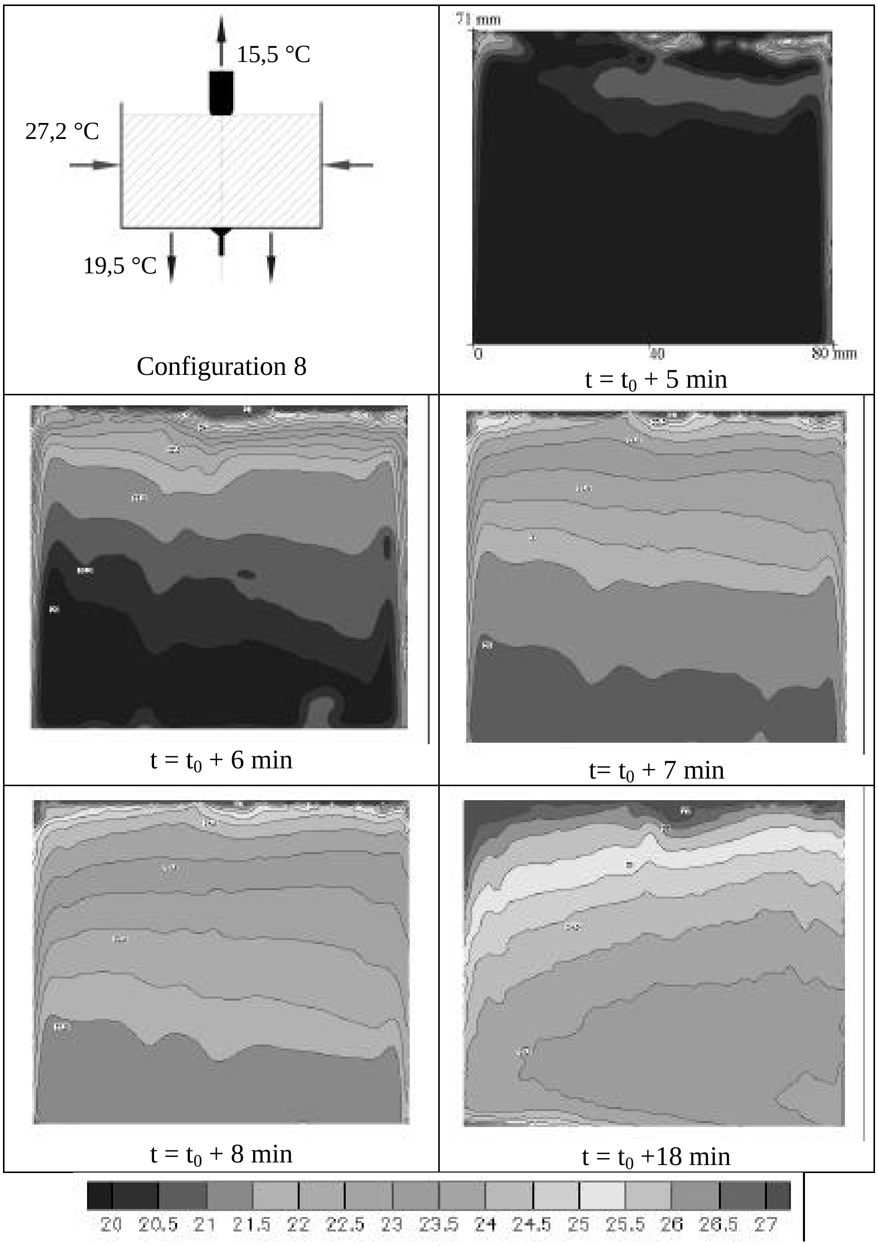

Figure 5: Qualitative analysis of transient isotherm progressions

Depending on the combination of different settings of thermal, kinematic and geometrical boundary conditions, there are 35 configurations possible. For each of them the transient isotherm progressions are recorded and the temperature and velocity fields have are quantitatively evaluated. The experiments were made with an 85 % and a 25 % glycerine-water mixture and with 4 different types of liquid crystals used as tracers.

The example in Figure 6 shows configuration 8. The experimental fluid is a 25 % glycerine-water mixture (Pr = 16.1) with the volume rate of 0.1 % liquid crystals, type KXN 2030 (Japan Capsular Products).

As can be seen from the qualitatively analysed temperature fields images (Figure 5), the temperature fields are clearly unsymmetrical. This is because of the large geometry of the measurement cubicle, with a diameter of 80 mm and a filling level of 71 mm.

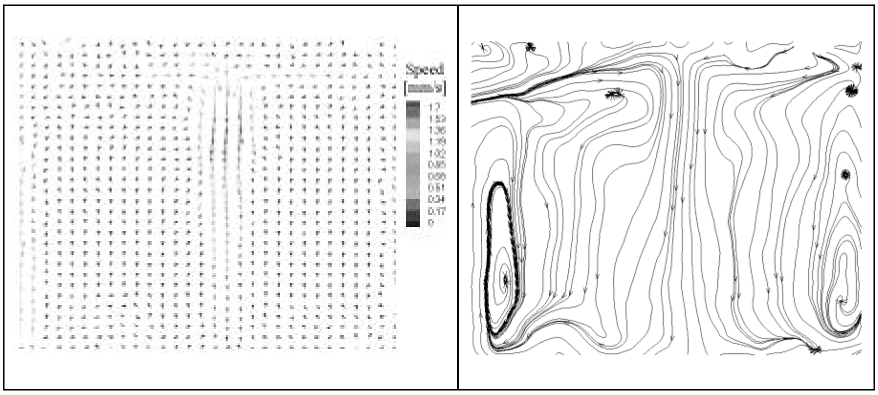

Figure 6: Velocity field and streamlines

As soon as the radial temperature gradient appears on the fluid surface between the wall and the middle of the measurement cubicle, the thermocapillary (Marangoni) convection also appears (Figure 6). That means that, in contrast to the Reynolds Number, no critical Marangoni Number exists, which determines the appearance of the thermocapillary convection. At this place a tangential tension appears, which causes the fluid to move towards the increasing interface tension from the edge to the middle of the cubicle. The flow thus created is transmitted by friction to deeper lying fluid layers. Due to reasons of continuity a closed convection roll will be formed. This causes a destruction of the homogeneous stable density layers in the volume and a secondary temperature gradient is generated, which is one reason of the gravitation in the volume caused by convection. When the measurement cubicle is adjusted, the forced convection also appears in the volume

The flow is laminar with several convection rolls which do not appear symmetrically to the vertical axis. There could be two reasons for the broken symmetry of the velocity fields: the velocity fields are only symmetrical until a value of the temperature gradient of dT/dr is reached or the secondary temperature gradient in the volume destroys the rotational symmetry. These two suppositions have still to be proved.

6 Perspectives and plans

The experimental equipment allows simultaneous recording and analysis of velocity and temperature fields in the Czochralski simulation. Its flexibility offers the opportunity to change many kinematic, thermal and geometrical factors and to investigate the influence of boundary conditions created in this way in order to optimise the process.

The method, the algorithm and the software for the qualitative analysis of the temperature fields have been developed and used for the Czochralski configuration.

In the future the topography of the flow and the interaction of the three types of convection, as well as the coupling between the velocity and temperature fields will be investigated. The oscillatory and turbulent Marangoni convection and the phase behaviour of velocity and temperature oscillations will be analysed, too. Moreover we will analyse the appearance of the oscillatory and turbulent Marangoni convection for low viscosity fluids.

Acknowledgement

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