Nonstationary melt flow and mixed convection in Czochralski process

V S Berdnikov, V V Vinokurov and V A Vinokurov
Kutateladze Institute of Thermophysics SB RAS, Novosibirsk, Russia

E-mail: berdnikov@itp.nsc.ru

Abstract. Regimes of mixed convection of melts with Prandtl numbers \( \text{Pr} = 10.78 \) and 16 were simulated experimentally and numerically in an application to hydrodynamic model of Czochralski with a fixed crucible case. In mixed nonstationary regimes the forced flow is induced by a uniformly rotating crystal under a given mode of thermal gravitational-capillary convection. The experiments and calculations were carried out with 96% ethanol (\( \text{Pr} = 16 \)) and water (\( \text{Pr} = 10.78 \)) as modelling fluids. When \( \text{Pr} = 10.78 \), abnormally high value of surface tension leads to an increase of the influence of the thermocapillary effect and the appearance of heat exchange features. Nonstationary flow regimes are determined within the range of Reynolds number \( 82 \leq \text{Re} \leq 103 \) at \( \text{Pr} = 16 \).

1. Introduction

Czochralski process is the main method of crystal growth used to obtain single crystals of semiconductors (e.g. silicon, germanium and gallium arsenide) from melts [1, 2]. This method is multivariate and modified depending on the thermophysical properties of melts and crystals. The need for a deep and complete study of thermophysical processes in the growing crystals objectively is due to emerging new crystalline materials and strict requirements for their quality. In real technology it is very difficult to study the hydrodynamic and complex conjugate heat transfer in melts in the crucible. The combination of experimental and numerical simulations is the most effective method of studying the model of Czochralski crystal growth.

In the Czochralski process, natural convection depends on the various temperature differences between the lateral crucible wall and the disk modelling the crystal, which is fundamentally unavoidable and poorly controlled [1–4, 6, 9]. The simplest way to control the convection and convective heat transfer is to rotate the crystal. The heat transfer in hydrodynamic model of Czochralski crystal growth was investigated experimentally [2, 4]. The influence of convective heat transfer on the shape of crystallization front with \( \text{Pr} = 45.6 \) [4] is studied experimentally. The relative role of buoyancy forces and the thermocapillary effects in technologically optimal mixed convection regimes has been poorly studied so far. This investigation is a continuation of the experimental and numerical simulations presented in [1–4, 6, 9]. The laminar thermal gravitational-capillary convection was numerically studied. At the given Gr and Ma a uniform rotation of the crystal was applied and the evolution of the flow structure and heat exchange with the increase in Reynolds number at \( \text{Pr} = 10.78, 16 \) was investigated. Mixed convection regimes can be stationary and non-stationary subject to relationships of Gr, Ma, and Re numbers. Numerical studies were performed using a finite difference
2. Problem definition

Numerical simulations were based on Navier–Stokes equations for axisymmetrical cases. The process in the crucible–melt-crystal system is a convection induced by cooled and uniformly rotating crystal, which partially covers the free melt surface. A stationary cylindrical container with heated sidewalls is used. The system of mixed convection equations in the Boussinesq approximation in terms of vorticity, stream function, temperature, and azimuthal velocity is solved:

\[
\frac{\partial \omega}{\partial t} + U \frac{\partial \omega}{\partial r} + V \frac{\partial \omega}{\partial z} - \frac{U}{r} = \frac{1}{Re} \left( \Delta \omega - \frac{\omega}{r^2} \right) - \frac{Gr \Delta \theta}{Re^2 \frac{\partial}{\partial r}},
\]

\[
\Delta \psi = 2 \frac{\partial \psi}{\partial r} = \rho \alpha,
\]

\[
\frac{\partial \theta}{\partial t} + U \frac{\partial \theta}{\partial r} + V \frac{\partial \theta}{\partial z} = \frac{1}{Pr Re} \Delta \theta,
\]

\[
\frac{\partial W}{\partial t} + U \frac{\partial W}{\partial r} + V \frac{\partial W}{\partial z} + \frac{U W}{r} = \frac{1}{Re} \left( \Delta \omega - \frac{W}{r^2} \right)
\]

The system of equations is supplemented with the following boundary conditions. The bottom of the crucible (rigid and adiabatic):

\[
\psi = 0, \quad \frac{\partial \psi}{\partial z} = 0, \quad \frac{\partial \theta}{\partial z} = 0, \quad W = 0, \quad z = 0, \quad 0 \leq r \leq R_T,
\]

the lateral surface of the crucible:

\[
\psi = 0, \quad \frac{\partial \psi}{\partial r} = 0, \quad \theta = 1, \quad W = 0, \quad 0 \leq z \leq H, \quad r = R_T,
\]

free melt surface (taking into account the thermocapillary effect on the thermally insulated boundary):

\[
\psi = 0, \quad \frac{\partial \psi}{\partial z} = 0, \quad \frac{\partial \omega}{\partial z} = 0, \quad W = 0, \quad z = H, \quad 1 \leq r \leq R_T,
\]

crystallization front:

\[
\psi = 0, \quad \frac{\partial \psi}{\partial z} = 0, \quad \frac{\partial \theta}{\partial z} = 0, \quad W = r, \quad z = H, \quad 0 \leq r \leq 1,
\]

the axis of symmetry:

\[
\psi = 0, \quad \frac{\partial \psi}{\partial r} = 0, \quad W = 0, \quad 0 \leq z \leq H, \quad r = 0.
\]

Dimensionless form of the equations uses the following parameters: the scale length is the radius of the crystal - $R_K$; temperature scale is the temperature difference between the edge of the crystal and the walls of the crucible – $\Delta T$; velocity scale is the linear speed of the edge of the crystal – $\omega K R$. Numerical simulation by finite difference method was carried out on uniform grids 160x160 or 320x320.

There are groups of dimensionless numbers in the system: the Prandtl number ($Pr$), defined as the ratio of momentum diffusivity to thermal diffusivity, the Grashof number (Gr), which approximates the ratio of the buoyancy to viscous force acting on a fluid, the Marangoni number (Ma) regarded as proportional to surface tension forces divided by viscous forces, the Reynolds number (Re) defined as dimensionless number of the crystal rotation rate:

\[
Gr = \frac{g \beta}{v^2} \Delta T \cdot R_K^3, \quad Ma = \left( \frac{\partial \sigma}{\partial T} \right) R_K \Delta T, \quad Pr = \frac{\nu}{\alpha}, \quad Re = \frac{\Omega K \cdot R_K^2}{\nu}.
\]
Figure 1. Isolines of stream function (L) and isotherms (R) at $Pr = 16$, $Re = 90$, $Gr = 5835$, $Ma = 4870$: a) $t = 244$ s, b) $295$ s, c) $310$ s, d) $388$ s.
3. Results and Discussion

Local characteristics in mixed convection result from interaction of buoyancy and thermocapillary driven convection and the flow induced by a uniformly rotating crystal. It was established experimentally [3-5] that when a critical Re depending on Gr and Ma is reached, a vortex appears under the rotating crystal. Laminar, stationary and axisymmetric melt flow occurs at Gr = 5835, Ma = 4870, 0 ≤ Re ≤ 81, 104 ≤ Re ≤ 500. Within the range 82 ≤ Re ≤ 103 the flow loses stability and axisymmetric vibrations occur.

Figure 1 shows the flow structure for different time instants for Gr = 5835, Ma = 4870 within the range 0 ≤ Re ≤ 81 and 104 ≤ Re ≤ 500. Each flow structure corresponds to the extreme values of Nusselt numbers:

\[ Nu = \left( \int_0^{2\pi} \int_0^{2\pi} (\partial \theta / \partial z)_{\theta = \int} r dr d\phi \right) / Q_{\lambda} \]

These values of Nusselt numbers represent the integrals of instantaneous radial distributions of dimensionless local heat fluxes shown in figure 2. Here Q_{\lambda} is integral heat flux on the crystallization front in the thermal conductivity mode. The analysis of the results in figures 1-3 shows that the Nu values are relatively low when the crystallization front is blocked by a portion of cold melt from the hot melt flow moving from the crucible walls (figure 1). At times corresponding to the maximum values of Nu, the cold melt is ejected from the crystallization front. Within nonstationary flow regimes (82 ≤ Re ≤ 95) temperature and local heat flux (figures 2) fluctuations make the main contribution to the Nu oscillations at the crystallization front.

The calculations were carried out using a water (Pr = 10.78) as modelling fluid which was chosen due to the high value of the surface tension coefficient and its significant dependence on temperature. The properties of water are taken at a temperature T = 6°C. Results at three values of Grashof and Marangoni numbers at Pr = 10.78, H/R_T = 0.7, R_T/K = 2.76 are shown in figure 4. Dependence Nu vs Re changes in a similar fashion for Re ≥ 200 at the qualitative level.

Unlike ethanol (Pr = 16), the relative role of the thermocapillary effect compared with buoyancy forces increases dramatically. It is clear that the Nusselt number varies abruptly at Re = 330 and Re = 550 at Gr = 924 and Ma = 21122 (corresponding ΔT = 2.0°C). This is a consequence of the rearrangement of the spatial shape of the melt flow and local heat fluxes at the crystallization front.

Experimental simulations using an ethanol as modeling fluid are presented in figure 5. The main regularities in the flow structure with increasing the Reynolds number practically coincide with our numerical simulations at Pr = 16. Experimental simulations and numerical simulations are carried out...
for identical geometries and dimensionless numbers (Gr, Ma, Re, Pr). The main difference between the experimental and the numerical simulations is that the flow acquires a three-dimensional character. The boundary between the flow induced by centrifugal forces and the flow induced by buoyancy forces loses stability. However, the qualitative data obtained from experimental and numerical simulations are similar.

4. Conclusions
Experimental and numerical simulations of the flow structure and heat exchange at fixed Prandtl, Grashof, and Marangoni numbers and increasing Reynolds number were carried out. Nonstationary flow regimes are determined at a fixed temperature difference between the crystal and crucible and at $\Pr = 16$ (ethanol as modelling fluid), $82 \leq Re \leq 103$. The melt flow loses stability that results in occurrence of axisymmetric oscillations. The grown Nusselt number varies abruptly with increasing of Reynolds number at $\Pr = 16$, $\Delta T = 2.0^\circ C$. A qualitative coincidence between the numerical and experimental data is obtained. In the case of $\Pr = 10.78$ the Nusselt number (Nu) changes nonmonotonically with increasing Re for sufficiently intense buoyancy and thermocapillary driven convection.

![Figure 4. Dependences of the dimensionless heat transfer coefficient vs the Reynolds number at $\Pr = 10.78$, $H/R_1 = 0.7$, $R_\gamma/R_K = 2.76$: 1 – $Gr = 230$, $Ma = 5280$; 2 – $Gr = 462$, $Ma = 10561$; 3 – $Gr = 924$, $Ma = 21122$.](image-url)
Figure 5. The flow structure at $Pr = 16$, $Gr = 5835$, $Ma = 4870$, $Re = 95$, $H/RT = 0.7$, $R_T/R_K = 2.76$

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