Features of convective heat transfer in mixed convection regimes in the Czochralski method with different effects of buoyancy forces and thermocapillary effect

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Abstract. The influence of the height of the melt layer on the boundary of the transition from stationary flow and heat exchange to non-stationary regimes for given values of Grashof, Marangoni and Reynolds numbers is investigated. The hydrodynamics, local and integral heat transfer were studied at the value of the Prandtl number 16. At the values of the Prandtl number 10.78; 16 and 45.6, integral heat transfer was studied at a fixed height of the melt layer with increasing Reynolds numbers.

1. Introduction

The Czochralski method produces single crystals of various materials and different sizes [1, 2]. This method has many variants of the growth site apparatus and is modified when growing materials with different thermal properties [1-3]. But in all cases in the Czochralski method, the crystal is drawn out from the free surface of the melt. Due to the temperature difference between the crystallization front and the walls of the crucible and the presence of the free surface of the melt, a fundamentally irremovable and poorly controlled thermal gravity-capillary convection occurs. In thermal gravity-capillary convection regimes, the melt flow can have a complex spatial shape and can become unsteady with increasing temperature drops and crystal sizes. Spatio-temporal characteristics of the flow depend not only on the values of the dimensionless similarity criteria: Grashof numbers $Gr = (\beta g/\nu^2)\Delta T R_k^3$ and Marangoni Ma $= [(d\sigma/dT)/\mu]\Delta T R_k$, but also on their relationship [3]. To control the hydrodynamics and convective heat exchange in the “crucible-melt-crystal” systems, the existing technologies use the selection of the crystal rotation speed (or Reynolds number $Re = \omega R_k^2/\nu$) [3–5]. This is accompanied by a transition from the initial free convection mode to mixed convection modes. For given Prandtl numbers of the melts $Pr = \nu/a$ and given geometrical parameters, relative radius $R_t/R_k$ and relative height of the melt layer $H/R_t$, the flow and convective heat exchange characteristics are already determined by the ratios of numbers $Gr$, $Ma$, $Re$ [3–5]. With increasing the angular velocity of rotation of the crystal, the contribution of centrifugal forces to the formation of the melt flow increases and the spatial flow patterns and flow modes of the crystallization front change. Accordingly, the conditions of conjugate convective heat transfer on the crystallization fronts and their shape change [5]. The need to control technological processes requires an understanding of how the modes of melt flow change in the bulk and in the boundary layer at the crystallization front (CF) with the changes in characteristic temperature drops and angular velocities of rotation of the crystals. Under process conditions, it is almost impossible to investigate the hydrodynamics of melts and heat transfer and purposefully select technologically optimal modes. The
most effective method for studying the processes under consideration is a combination of physical and numerical modelling.

In the classical version of the method with a fixed crucible, as the crystal grows, the level of the melt decreases. With a decrease in the melt level, the relative role of the buoyancy forces and the thermocapillary effect in the processes of generating convective flow changes with formally the same values of the Gr and Ma numbers. Therefore, the flow characteristics at a fixed relative radius \( R_T/R_K \) depend on the relative height of the melt layer \( H/R_T \). The influence of these parameters is not fully understood and is the subject of research in this work. In [5], the transition from laminar stationary modes of mixed convection to nonstationary periodic and quasiperiodic flow modes was found at \( R_T/R_K = 2.76 \) and \( H/R_T = 0.7 \) in the range of Reynolds numbers \( 82 \leq Re \leq 105 \) at \( Gr = 5835, Ma = 4870, Pr = 16. \) Below are the results obtained with the same values of the Gr and Ma numbers, with \( Re = 95, \) but in a wide range of \( H/R_T. \)

2. Formulation of the problem

The dependence of hydrodynamics and heat transfer on the relative height of the fluid layer in mixed convection modes with a constant Reynolds number and for given values of Grashof and Marangoni numbers was studied numerically using the finite-difference method. The scheme of the computational domain is shown in Fig. 1. Here the cold crystal model with a flat front \( S4 \) (at temperature \( T1 \)) located at the free surface of the melt \( S3 \) has radius \( R_K, \) a crucible of regular cylindrical shape has radius \( R_T, \) isothermally heated to \( T2, \) side walls \( S2 \) and insulated bottom \( S1. \) The crucible is filled with a melt to the level \( z = H \) with a given Prandtl number (region \( \Omega). \) So, the side walls of the crucible are set by isothermal heating, the model crystallization front is isothermally cold, the bottom and the free surface from the edge of the crystal to the walls of the crucible are adiabatic. The crystal model rotates uniformly with a given angular velocity \( \Omega_K. \)

The original system of dimensionless equations of mixed convection in the Boussinesq approximation and assuming axial symmetry of the velocity and temperature fields in variables (vortex, stream function, temperature, azimuth velocity) had the form:

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

\[
\Delta \psi - \frac{2}{r} \frac{\partial \psi}{\partial r} = r \omega;
\]

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

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

In the transition to the dimensionless form of the equations and boundary conditions, the following scales are used as scales: length scale is crystal radius \( R_K; \) temperature difference between the crystal edge and the crucible walls is \( \Delta T = T_2 - T_1; \) speeds are represented by linear speeds of crystal edge \( \Omega_K R_K; \) time scale is \( 1/\Omega_K. \) The system includes four similarity criteria: the Grashof number \( Gr, \) which determines intensity of thermogravitational convection; Marangoni number \( Ma, \) characterizing the contribution of the thermocapillary effect; Prandtl number \( Pr \) characterizing the thermophysical
properties of the melt and the Reynolds number $Re$, which determines the intensity of forced convection:

$$Gr = \frac{g \beta}{v^2} \Delta T \cdot R_K^3, \quad Ma = \left( \frac{\partial \sigma}{\partial T} \right)_{a \mu} \cdot R_K \cdot \Delta T, \quad Pr = \frac{v}{a}, \quad Re = \frac{\Omega_K \cdot R_K^2}{v}.$$  

Here $\sigma$ is the coefficient of surface tension, $\mu, v$ are the coefficients of dynamic and kinematic viscosity, $a$ is the coefficient of thermal diffusivity. When analyzing the results and just the physical essence of the processes in mixed convection, another parameter is important, sometimes called the Richards number $Ri = Gr/Re^2$. This parameter is a measure of the relative role of buoyancy forces and inertia forces.

Mathematical record of conditions for all elements of the boundary has the form:

S1. Crucible bottom (rigid, adiabatic):

$$\psi = 0, \quad \frac{\partial \psi}{\partial z} = 0, \quad W = 0, \quad \frac{\partial \theta}{\partial z} = 0, \quad z = 0, \quad 0 \leq r \leq R_T/R_K;$$

S2. Side surface of a cylindrical fixed crucible:

$$\psi = 0, \quad \frac{\partial \psi}{\partial r} = 0, \quad W = 0, \quad \theta = 1, \quad 0 \leq z \leq H/R_K, \quad r = R_T/R_K;$$

S3. Fluid free surface (adiabatic):

$$\psi = 0, \quad \omega = -\frac{Ma}{Pr \cdot Re} \frac{\partial \theta}{\partial r}, \quad \frac{\partial W}{\partial z} = 0, \quad \frac{\partial \theta}{\partial z} = 0, \quad z = H/R_K, \quad 1 \leq r \leq R_T/R_K;$$

S4. Crystallization front:

$$\psi = 0, \quad \frac{\partial \psi}{\partial z} = 0, \quad W = r, \quad \theta = 0, \quad z = H/R_K, \quad 0 \leq r \leq 1;$$

S5. Symmetry axis:

$$\psi = 0, \quad \omega = 0, \quad W = 0, \quad \frac{\partial \theta}{\partial r} = 0, \quad 0 \leq z \leq H/R_K, \quad r = 0;$$

Numerical simulation by the finite difference method was carried out mainly on a uniform 160x160 or 320x320 grid. The dependence of hydrodynamics and heat transfer on the relative height of the fluid layer with the Prandtl number $Pr = 16$ was investigated with fixed values of the remaining parameters $Gr = 5835$, $Ma = 4870$, $Re = 95$, $R_T/R_K = 2.76$.

3. Results and discussion

The main result of the calculations is presented in Figure 2 and consists in the fact that in the range of $0.2 \leq H/R_T \leq 0.35$, the flow regime is stationary. And starting from $H/R_T = 0.36$, oscillations of a quasi-periodic nature develop. In Figure 2, curve 1 shows the dependence of the integral of the heat transfer coefficient (Nusselt number $Nu$) on the height of the liquid layer, obtained by integrating the radial distributions of local heat fluxes (Figure 3) over the area and time. Figure 4 shows the time dependence of the integral heat transfer coefficient, determined by integration of $Nu_1$ over the area of the crystallization front at different time points. The integration time interval for determining $Nu$ corresponded to several low-frequency oscillations of $Nu_1$ clearly visible in Fig. 4. Curve 2 in Fig. 2 shows the dependence of $Nu$ on $H/R_T$ in stationary flow regimes.

An analysis of the data presented in Figures 5-13 shows how the transition from stationary mode to oscillations occurs. The evolution of the spatial shape of the flow and temperature fields with increasing height of the liquid layer in stationary regimes is shown in Figure 5.
It is seen that the flow of hot liquid pushes the lower part of the cocurrent descending flow from the edge of the crystal to the bottom of the crucible to the axis. Here and below, only the right-hand part of the symmetric spatial form of the flow and fields of isotherms is shown. Distributions of local heat fluxes and profiles of the vertical velocity component are shown in Figures 6 and 7. The transition to relatively weak axisymmetric oscillations can be seen in Figure 8 for changes in the shape and position of the similar isolines of the stream function over a period of time Δt = 47.9 s.

These oscillations are accompanied by temperature pulsations and oscillations of local heat fluxes at the crystallization front. Figure 3 shows in close-up only the central part of the radial distributions of local heat fluxes q (r, t) at different points in time.

The maximum value of q (r, t) at the crystal edge is q_{max} (r, t) = 42.9. Oscillations have small amplitude, but they are low-frequency and, therefore, their appearance may be the cause of the banded heterogeneity of crystals. To clarify the nature of the oscillations, phase trajectories of oscillations of the temperature difference between two points in the fluid layer were constructed. The first reference point is 10 steps down from the crystallization front and 10 steps from the front edge to the symmetry axis by 3 grid steps 160 × 160. The second point is at the same height level and is removed by 10 grid steps in the radial direction from the first point. It is located at a distance of 7 grid steps behind the edge of the crystallization front. A confluent downstream is in between. The phase trajectories in Figure 9 show the element quasi-periodicities that become more apparent if you select individual fragments for shorter periods of time. They are shown in Figure 9.
Figure 5. Fields of isolines of the stream function and isotherms.

Figure 6. Radial distributions of local heat fluxes.

Figure 7. Profiles of the vertical velocity component in the section $z = \frac{H}{2}$. 

$H/RT = 0.20$

$H/RT = 0.35$, $Pr = 16.0$

$Pr = 16$, $1 - H/RT = 0.2$, $2 - 0.31$, $3 - 0.35$

$Pr = 16; 1 - H/RT = 0.2$, $2 - 0.31$, $3 - 0.35$;
Here the dots show the beginning of the trajectories. Over a period of more than three quasi-periods of low-frequency oscillations of the Nu1 number (Figure 4), the trajectories do not close. That is, the oscillations have a stochastic quasi-periodic character, and not a complicated periodic one. With an increase in the height of the liquid layer, the amplitudes of oscillations of the downhole companion flow, temperature, and local heat flow increase (Figures 10-12). The instantaneous fields of isotherms and isolines of the stream function presented in Figure 10 make it possible to understand the causes of fluctuations in heat fluxes. They consist in a quasi-periodic process of leakage of the heated liquid under the crystallization front and emissions of the cooled liquid to the periphery.

The quasi-periodicity of the process is shown by the data in Figure 10 and Figure 12. Figure 10 shows the moments of almost repetition of the forms of isolines of the stream function and isotherms in about 100 seconds. Similar data were obtained for high layer heights.

Figures 11 and 12 present data on the dependences of local and integral characteristics at the height of the layer H/RT = 0.4. Figure 12 shows the dependence of the average FC area of the heat flux on time. The average values are obtained by integrating instantaneous values of local heat fluxes over the area of the FC (Figure 11).

The relative role of the buoyancy forces and the thermocapillary effect can be estimated from the temperature and velocity distributions along the free surface of the melt and from the profiles of the radial velocity component, i.e. along the distribution of the height of the layer in given sections along the radius (Fig. 13).

The presented data were obtained for a given same temperature difference between the crystal model and the crucible walls and for fixed values of Grashof and Marangoni numbers, in which the characteristic size is the radius of the crystal. The Reynolds number is also fixed.
In the height range of the layer $0.2 \leq H/RT \leq 0.4$, the collision boundary of the free convective and forced flows is in the subcrystal region near the crystal edge practically not moving in the radial direction.

Figure 11. Radial distributions of local heat fluxes at different points in time: 1 – 71.3 s; 2 – 114.3; 3 – 138.0; 4 – 146.5; 5 – 154.4; 6 – 175.5 s.

Figure 12. Time dependence of the average FC area of the dimensionless heat flux at $H/RT = 0.40$, $Pr = 16$.

Figure 13. Radial distributions of temperature (a) and velocity (b) on the free surface of the melt: 1 – $H/RT = 0.2$, 2 – $H/RT = 0.31$, 3 – $H/RT = 0.35$, 4 – $H/RT = 0.36$, 5 – $H/RT = 0.4$. 
direction when going to the unsteady flow mode at $H/RT = 0.36$. In Fig. 13a, it can be seen that at $H/RT = 0.2$, the temperature gradient along the free surface has a maximum value and the thermocapillary effect makes a significant contribution to the flow generation in the region from the walls of the crucible to the edge of the crystal. The flow rate along the free surface to the crystal edge has a maximum value (Figure 13b). The profiles of the radial velocity component in Fig. 13c also show that at $H/RT = 0.2$ the influence of the thermocapillary effect on the flow is significant.

With an increase in the height of the liquid layer, the influence of the thermocapillary effect in the central part decreases sharply against the background of thermogravitational convection. But near the edge of the crystal and at the walls of the crucible, the temperature gradients along the free surface remain significant. At the edge of the crystal, they even increase.

The observed patterns were verified experimentally on the physical model of the Czochralski method. A videotaping of the melt flow of heptadecane in mixed convection modes with fixed temperature difference and angular velocity of rotation of the crystal model with a discrete set of heights of the liquid layer was performed. The results confirm the numerically detected patterns. Figure 14 shows the trajectories of the visualized fluid at two heights of the fluid layer.

The results of numerical studies obtained for three liquids - simulators of melts: ethyl alcohol $Pr = 16$, water $Pr = 10.78$ and heptadecane $Pr = 45.6$, are summarized and partially presented in Figure 15 as dependences of dimensionless heat transfer coefficients - Nusselt numbers on the Reynolds number. At $Pr = 16$ and 10.78, data were obtained at $H/RT = 0.7$ and $RT/RK = 2.76$, and at $Pr = 45.6$, with the same height of the liquid layer, but with a larger diameter FC. The main features in Nu ($Re$) dependencies are: relatively high Nu values in starting modes of free convection decrease monotonously when turning on the crystallization front and reach minimum values when a centrifugal vortex forming the entire crystallization front area is formed under the rotating crystallization front. In this case, the crystallization front is blocked by a cooled liquid from a stream of heated liquid that flows onto the edge of the crystallization front from heated side walls. As the intensity of forced convection increases and the current of free convective nature pushes aside, Nu ($Re$) begins to increase monotonously due to more efficient heat transfer on the crucible walls and with convective flow to the crystallization front. The interaction area of the cold flow from the rotating FK and the flow from the crucible walls increases monotonically. In

![Figure 13 c. The profiles of the radial velocity component in the section $r = (R_x + R_h)/2$ with $R_x/R_h = 2.76$: 1 – $H/RT = 0.2$, 2 – $H/RT = 0.31$, 3 – $H/RT = 0.35$, 4 – $H/RT = 0.36$, 5 – $H/RT = 0.4$.](image1)

![Figure 14. Motion trajectories: $Pr = 45.58$, $Gr = 8900$, $Ma = 5716$, $Re = 75.4$.](image2)
addition, the crucible walls begin to be flown around by an increasingly swirling flow. In the case of heptadecane, a hysteresis effect is observed in the modes of monotonous change in the angular velocities of rotation of the crystals, as can be seen from curve 5 in Figure 15. Its presence is confirmed by calculations on grids of higher dimension. The growth of the Nu number on the upper branch is explained by the centrifugal flow of the crucible walls, and in flow regimes corresponding to the lower branch of the crucible wall, it is blocked by free convective flows.

Conclusion
The influence of the height of the melt layer on the boundary of the transition from stationary flow and heat transfer to unsteady flow regimes for given values of Grashof, Marangoni and Reynolds numbers is studied numerically. Numerical studies are performed by the finite difference method (using proprietary software). In the stationary and non-stationary mixed convection modes, hydrodynamics, local and integral heat transfer are studied at a Prandtl number of 16. The conclusions about the presence of the transition boundary to oscillations with increasing melt layer height were tested experimentally on a fluid with Pr = 45.6. With the values of Prandtl numbers 10.78; 16 and 45.6, the integral heat transfer was studied at a fixed height of the melt layer with increasing Reynolds numbers. The calculation results for fixed values of the Gr and Ma numbers are summarized as dependences of the Nusselt number Nu (Re).

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