Numerical simulation of crystallization on a stationary and rotating cooled disk

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Abstract. The process of crystallization of silicon melt on a monotonically cooled disk located on the free surface of the melt is studied numerically. The influence of thermogravitational, thermal gravitational-capillary and mixed convection on the shape of the crystal–melt interface is investigated. In mixed convection modes, the speed of uniform rotation of the disk is set. The calculations were carried out in an axisymmetric formulation of problems by the finite element method using an adaptive triangular mesh and taking into account the latent heat.

1. Introduction
Crystallization from melts occurs in systems that are inevitably non-isothermal and in a gravity field. Therefore, thermogravitational convection develops in melts [1-3]. In the presence of a free nonisothermal surface of the melt, the action of the thermocapillary effect (TCE) is added and convection has a thermal gravitational-capillary nature [1-3]. The spatial shape and intensity of flows depend on the characteristic temperature drops, geometry and absolute dimensions of the working section of the growth apparatus. In the Czochralski and Kyropoulos methods, crystals are pulled from the free surface of the melt layers [1-3]. In works dealing with the analysis of the processes of obtaining single crystals by directed crystallization methods, it is emphasized that free convection is poorly controlled. The Czochralski method uses crystal rotation to control the hydrodynamics of melts. In this case, the physical nature of convective motion is gravitational-centrifugal [2-4]. Despite a large number of studies of directed crystallization methods, due to the nonlinearity of the interaction of various flow generation mechanisms, there are no answers to many questions. Until now, there are few works in which convective heat transfer is studied for cases when the forces of buoyancy, TCE, and rotation effects act [4]. An important, but insufficiently studied stage is the initial stage of crystal growth. Technological practice shows that by choosing the mode of convective heat transfer, it is possible to correct the shape of the crystal-melt interface (CMI). The development of technologies for growing high-quality single crystals requires an understanding of the features of conjugate convective heat transfer for various combinations of thermal conductivity of melts, crystals, and crucible materials. This work is aimed at investigating this influence. For the silicon-graphite system, these studies are the development of works [2, 3]. Direct experimental investigations of high-temperature technological processes are expensive and laborious. It is practically impossible to measure the characteristics of non-stationary temperature fields in the composite region of the crucible-melt-crystal of the growth apparatus. Therefore, it is advisable to numerically investigate the effect of conjugate convective heat transfer on the crystallization process and on the forms of CMI.
2. Statement of the problem

The problem statement corresponds to a simplified model of the initial stages of crystal growth in the Kyropoulos and Czochralski methods. In these methods, crystals are grown by pulling them out of the melt layer with a free surface [1]. Crystallization of a silicon melt in a graphite crucible is investigated. The crucible is a cylindrical container made of MPG-6 graphite with a wall and bottom thickness of 0.5 cm. The inner radius of the crucible is 5 cm. The height of the melt layer is 5 cm. The crystallization process begins on a monotonically cooled disk located at the upper boundary of the melt. The radius of the disc is 2 cm. The problems were solved in an axisymmetric setting in a composite computational domain, the diagram of which is shown in figure 1. Only the right side of the axisymmetric computational domain is shown here. At the initial moment of time \( t = 0 \), the crucible (\( \Omega_1 \)) is filled with melt (\( \Omega_2 \)), overheated relative to the crystallization temperature (1410 °C) by 10 K. The initial melt temperature is maintained on the outer vertical surface of crucible \( \Omega_5 \). The disk is cooled by lowering the temperature at the \( S_{10} \) boundary at a rate of 2 K/min. The crystallization process begins when the temperature on the disk drops to the crystallization temperature. In the mixed convection mode, the angular velocity of uniform rotation of the disk is set (i.e., boundaries \( S_{10}, S_9 \), and \( S_{11} \)). The upper free boundary of the melt (\( S_3 \)) is set flat and non-deformable, either the frictionless condition is specified on it, or TCE is taken into account. The total volume of the crystallized substance (\( \Omega_2 \)) and the melt is assumed to be constant. Convective heat transfer in a melt is described by a system of dimensionless equations of unsteady thermogravitational convection in the Boussinesq approximation in the variables temperature, stream function, vortex of velocity and in a cylindrical coordinate system have the form:

\[
\begin{align*}
\frac{\partial T}{\partial t} + U \frac{\partial T}{\partial r} + V \frac{\partial T}{\partial z} &= \frac{1}{Pr} \nabla^2 T + Q, \\
\frac{\partial \omega}{\partial t} + U \frac{\partial \omega}{\partial r} + V \frac{\partial \omega}{\partial z} - U \frac{\omega}{r} - \frac{1}{r} \frac{\partial W^2}{\partial z} &= \nabla^2 \omega - \frac{g \cdot \beta_r(T) \cdot R^2 \Delta T}{r^2} \frac{\partial T}{\partial r}, \\
\frac{\partial W}{\partial t} + U \frac{\partial W}{\partial r} + V \frac{\partial W}{\partial z} + U \frac{W}{r} &= \nabla^2 W - \frac{W}{r^2}, \\
\nabla^2 \psi - \frac{2}{r} \frac{\partial \psi}{\partial r} &= r \cdot \omega, \quad U = \frac{1}{r} \frac{\partial \psi}{\partial z}, \quad V = -\frac{1}{r} \frac{\partial \psi}{\partial r},
\end{align*}
\]

\( U, V, W \) are the components of the velocity vector, \( T \) is temperature, \( \omega \) is a vortex of velocity, \( \psi \) is the stream function, \( \nabla^2 \equiv \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2} \) is Laplace operator, \( Pr = \nu / \rho \lambda / \lambda_f \) is Prandtl number, \( \Delta T \) is characteristic temperature difference. The dimensionless equations of heat conduction in the crystal and crucible have the following form, respectively:

\[
\begin{align*}
\frac{c_r \rho_s}{c_f \rho_f} \frac{\partial T}{\partial t} &= \frac{\lambda_s}{\lambda_f} \nabla^2 T + Q, \\
\frac{c_c \rho_s}{c_f \rho_f} \frac{\partial T}{\partial t} &= \frac{\lambda_c}{\lambda_f} \nabla^2 T.
\end{align*}
\]

In the equations being solved \( c_p, c_s, c_c, \lambda_f, \lambda_s, \lambda_c \) are the specific heat coefficients of the melt, crystal, and crucible, respectively, \( \rho_s, \rho_c, \rho_f \) are the densities, \( \nu_f \) is the kinematic viscosity of the melt, \( \beta_f \) is the volumetric thermal expansion coefficient of the melt, \( g \) is acceleration due to gravity, \( R_f \) is scale of geometric dimensions.

The latent heat is accounted for through an internal heat source. Let for a time instant equal to \( c \) the
crystal-melt interface changes its position from $B^0(t)$ to $B^1(t)$ (figure 2), respectively, the source power per unit volume in the domain $\Omega^0(t)$, bounded by the surfaces $B^0(t)$ and $B^1(t)$, is equal to $Q = \frac{\rho L}{\Theta}$. Thus, in equations (1) and (2) the value of $Q$ is defined as:

$$Q = \begin{cases} \frac{L \cdot R_f^2}{\Theta \cdot c_f \, \sqrt{\Delta T}}, & \text{in the area of } \Omega^0(t); \\ 0, & \text{in the areas of } \Omega^1(t) \text{ or } \Omega^2(t). \end{cases}$$

It is clear that the position of the crystal-melt interface $B^1(t)$ is not known in advance. It is determined by solving discrete analogs of equations, by their multiple solution within one time step with a relaxation coefficient, while the position of the boundary $B^0(t)$, corresponding to the isothermal surface with a temperature value equal to the crystallization temperature is redefined, and the position of the boundary $B^1(t)$ remains unchanged.

At the boundaries between the «crucible-melt», «crucible-crystal», «melt-crystal», the conditions of ideal thermal contact are fulfilled, i.e., at the indicated boundaries, the temperature and heat flux are inseparable:

$$\lambda_f \frac{\partial T}{\partial n} \bigg|_{S_{t+1}} = \lambda_s \frac{\partial T}{\partial n} \bigg|_{S_{t+2}},$$

$$\lambda_f \frac{\partial T}{\partial n} \bigg|_{S_{t-1}} = \lambda_c \frac{\partial T}{\partial n} \bigg|_{S_{t-2}} \quad \text{or} \quad \lambda_c \frac{\partial T}{\partial n} \bigg|_{S_{t+2}} = \lambda_s \frac{\partial T}{\partial n} \bigg|_{S_{t-2}},$$

$$T \big|_{S_{t-1}} = T \big|_{S_{t-2}}, \quad T \big|_{S_{t+1}} = T \big|_{S_{t+2}}, \quad T \big|_{S_{t+1}} = T \big|_{S_{t-2}}.$$  

At the non-deformable boundaries of the melt region and at the «melt-crystal» interface, the no-flow conditions are set:

$$\psi \big|_{S_i} = 0, \quad i = 1, 2, 3, 9, 10.$$  

On the inner rigid surfaces of the crucible walls and on the «melt-crystal» interface, the adhesion conditions are satisfied, as a result of which the condition for the velocity vortex is presented in the form:

$$\omega \big|_{S_i} = \frac{\partial U}{\partial z} - \frac{\partial V}{\partial r}, \quad i = 1, 2, 9, 10.$$  

If the upper boundary of the melt is free with no friction, then:

$$\omega \big|_{S_3} = 0.$$  

If TCE is taken into account at the upper free boundary, then:

$$\omega \big|_{S_3} = \frac{Ma}{Pr} \frac{\partial T}{\partial r}, \quad Ma = \frac{-\bar{\sigma} R_f c_f}{\bar{\sigma} v_f \lambda_f} \Delta T.$$  

The following conditions are set on the axis of symmetry:

$$\psi \big|_{r=0} = 0, \quad \omega \big|_{r=0} = 0, \quad W \big|_{r=0} = 0, \quad \frac{\partial V}{\partial r} \bigg|_{r=0} = 0, \quad \frac{\partial T}{\partial r} \bigg|_{r=0} = 0.$$  

The upper horizontal boundary of the computational domain outside the disk and the lower horizontal boundary are adiabatic:

$$\frac{\partial T}{\partial z} \bigg|_{S_i} = 0, \quad i = 3, 6, 8, 11.$
At the initial moment of time, the temperature in the entire system is constant and exceeds the crystallization temperature, and there is no convective flow:

\[ T|_{t=0} = T^*, \quad \psi|_{t=0} = 0, \quad \omega|_{t=0} = 0, \quad W|_{t=0} = 0. \]

The initial temperature of the system is maintained on the outer vertical side surface of the crucible:

\[ T|_{S_7} = T^*. \]

The temperature on the disk changes over time:

\[ T(t)|_{S_{10}} = T^* - \frac{dT}{dt} t, \]

where \( \frac{dT}{dt} \) is the specified disk cooling rate.

Azimuthal velocity at the disk and crystal surfaces \((S_9, S_{10})\) is a function of the radius:

\[ W|_{S_{9,10}} = rW^*, \]

where \( W^* \) is the dimensionless azimuthal velocity at \( r = 1 \).

The problems were solved by the finite element method [5]. To approximate the solution, linear functions on triangles were used. Mesh generation is based on the direct method of constructing the Delaunay triangulation over the largest angle with cellular acceleration. The grid is built for each time step, so as to track the shape of the CMI at the current step and to track the shape of the CMI at the next step with automatic determination of the volume of the crystallized substance. In addition, the grids are thickened to the CMI on both sides and to the boundaries of the calculated area.

The calculations were carried out at the values of thermophysical parameters characteristic of silicon crystallization in graphite crucibles [6-8]: thermal conductivity coefficients of silicon melt \( \lambda_f = 67 \ W/(m\cdot K) \), silicon crystal \( \lambda_c = 22 \ W/(m\cdot K) \), graphite \( \lambda_g = 50 \ W/(m\cdot K) \); density of silicon melt and silicon crystal \( \rho_f = \rho_c = 2530 \ kg/m^3 \), graphite \( \rho_g = 1800 \ kg/m^3 \); specific heat of graphite \( c_g = 2100 \ J/(kg\cdot K) \), silicon melt and crystal \( c_f = c_s = 1000 \ J/(kg\cdot K) \); kinematic viscosity of silicon melt \( \nu_f = 3.4 \cdot 10^{-5} \ m^2/s \); volumetric expansion coefficient of silicon melt \( \beta_f = 1.4 \cdot 10^{-4} \ 1/K \); latent heat of silicon melt \( L = 1.8 \cdot 10^6 \ J/kg \); surface tension dependence on silicon melt temperature \( \partial \sigma/\partial T = -10^3 \ J/(m^2\cdot K) \); Prandtl number of silicon melt \( Pr = 0.0128 \).

3. Results and discussion

In the modes of nonstationary thermogravitational convection, the crystallization of silicon melt on the surface of a monotonically cooled disk after reaching the crystallization temperature. Then, similar studies were carried out taking into account the TCE in the mode of thermal gravitation-capillary convection. So the influence of unsteady natural convection on the hydrodynamics of the melt, heat transfer, and the shape of the CMI, excited either only by buoyancy forces or by the combined influence of buoyancy forces and the TCE, has been studied. At the next stage, these modes of natural convection were the initial ones in the calculations of mixed convection. In mixed convection modes, a forced flow occurs when the cooled disk rotates uniformly.

Figures 3a and 4a show the combined fields of isotherms and isolines of the stream function in the modes of thermogravitational and thermal gravitation-capillary convection in the «silicon-graphite» system at the same times \( t = 750 \ s \). Figures 3b and 4b show the combined fields of isotherms and isolines of the stream function in mixed convection modes at an angular rotation rate of the cooled disk \( \omega_K = 20 \ rpm \) at the same time moments \( t = 750 \ s \). The step between the isolines of the stream function \( \psi_h \) in figures 3a and 4a is 10 \ mm²/s, in figures 3b and 4b is 5 \ mm²/s. Isolines of the stream function with a value of 0 \ mm²/s are excluded. Figures 3, 4 show only the right-hand sides of the axisymmetric fields of the stream function isolines and thermospheres. The directions and intensity of the flow can be understood from the data on the profiles of the radial and axial velocity components in figures 5 – 8. In the main large-scale vortex (figures 3 and 4), the circulation of the melt occurs counterclockwise. In all flow regimes along the bottom, a melt flow, cooled under the CMI, flows onto
Figure 3. Fields of isotherms and isolines of the stream function in the thermogravitational convection mode (a) and in the mixed convection mode (b).

Figure 4. Fields of isotherms and isolines of the stream function in the mode of thermal gravitational-capillary convection (a) and in the mode of mixed convection (b).

Figure 5. Profiles of the radial velocity component in the section $r = 3 \text{ cm}$ in the modes of thermal gravitational-capillary convection (1), thermogravitational convection (3) and mixed convection (2, 4).

Figure 6. Profiles of the radial velocity component in the section $r = 4 \text{ cm}$ in the modes of thermal gravitational-capillary convection (1), thermogravitational convection (3) and mixed convection (2, 4).

The heated melt floats upwards along the inner side of the side wall of the crucible, but not to the free surface of the melt. As can be seen in figures 3 and 4 by the shapes of the isotherms with $T = 1419 \, ^\circ\text{C}$, in the upper right corner the melt is slightly overheated and the main part of the ascending flow of the heated melt leaves the wall towards the center, without excessive buoyancy. Before the onset of crystallization, the flow along the free surface of the melt in free convection modes
is directed from the crucible wall to the disk edge, and then to the CMI edge (figure 7). In the mode of thermal gravitational-capillary convection, the speed along the free surface is significantly higher (figure 7). This is also seen in figures 5 and 6 from a comparison of the velocity values on the free surface of the melt. A feature of the near-surface sections of the profiles in figures 5, 6 is the absence of a velocity gradient in the thermogravitational convection mode and its presence in the thermal gravitational-capillary convection. This is the influence of the TCE in the presence of a longitudinal temperature gradient along the free surface of the melt (figure 9).

![Figure 7. Profiles of the radial velocity component in the modes of thermal gravitational-capillary convection (1) and thermogravitational convection (3) in the section z = 5.5 cm and mixed convection (2, 4).](image)

![Figure 8. Profiles of the axial velocity component in the modes of thermal gravitational-capillary convection (1) and thermogravitational convection (3) in the section z = 4.5 cm of mixed convection (2, 4).](image)

A significant effect of TCE is also observed in mixed convection modes. This is especially clearly seen from the velocity profiles along the free surface of the melt in figure 7. Disregarding the TCE, the flow along the free surface is directed from the rotating surface to the crucible walls. In this case, a forced centrifugal flow from under the rotating surface displaces or even suppresses the free convective flow of the heated melt from the crucible walls to the CMI edge.

A clockwise vortex forms under the free surface. This can be seen in figures 5, 6 (curves 4). When TCE is taken into account along the free surface, the flow direction in the mixed convection regimes remains the same as in the thermal gravitational-capillary convection regime.

![Figure 9. Temperature distributions in the modes of thermal gravitational-capillary convection (1) and thermogravitational convection (3) in the section z = 5.5 cm and mixed convection (2, 4).](image)

![Figure 10. Temperature distributions in the modes of thermal gravitational-capillary convection (1) and thermogravitational convection (3) in the cross section r = 3 cm of mixed convection (2, 4).](image)
That is, a stream of heated melt runs on the edge of the CMI. Figures 3b and 4b show that under the influence of TCE, the hot melt flows more efficiently under the CMI. These features also affect the temperature distribution over the height of the melt layer (figure 10, curve 4, and figures 3b and 4b). This leads to the dependence of the CMI shape (corresponding to the position of the isotherm with $T = 1410\,^\circ\text{C}$) on the flow regimes (figure 11). In figure 11 (curve 2), it can be seen that the TCE enhances the flow incident on the edge of the CMI and, as a result, the radius of the CMI near the free surface of the melt decreases relative to the position shown in figure 11 of curve 4. Curve 4, corresponding to the regime without taking into account the TCE, shows that the radius of the CMI is larger, since centrifugal forces push the heated melt flow away from the CMI edge. However, the CMI moved downward less, since the upward flow of the heated melt flowing onto the CMI is more uniform and more intense. This can be seen from a comparison of curves 2 and 4 in figure 8.

4. Conclusion
Numerical simulation of the initial stage of silicon crystallization on a monotonically cooled disk is carried out. The dependence of the shape of the crystal-melt interface on the heat transfer regime at a fixed time interval of 750 s after turning on the disk cooling was determined. The studies were carried out in non-stationary modes of thermogravitational, thermal gravitational-capillary and gravitational-centrifugal convection (at an angular speed of rotation of the disk of 20 rpm). The study was carried out at an initial overheating of the melt relative to the crystallization temperature by 10 K and at the rate of monotonic cooling of the disk of 2 K/min. It is shown that in unsteady modes of mixed convection with a disk radius of 2 cm and crystal rotation rates up to 20 rpm, the presence of the thermocapillary effect significantly affects the heat transfer and the shape of the crystallization front.

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