Radiation-convective heat transfer from the crystals in methods of pulling from melts

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Abstract. The radiation-convective heat transfer from the crystals in methods of pulling from melts such as Czochralski, Stepanov and floating zone is investigated numerically in the conjugate statement of the problem. The calculations are performed in the ranges of Grashof numbers from 1000 to 25000, with the gas Prandtl number equal to 0.68. Calculations of convective heat transfer are carried out by finite element method, and radiation fluxes are calculated by the zonal method. The relative role of thermal conductivity, convective heat transfer and radiation heat transfer is investigated.

1. Introduction
In widely used methods of crystal pulling from melts, the structural perfection of the crystals obtained largely depends on such parameters as the symmetry of temperature fields in the furnace, the rate of crystal growth, and the absence of sharp changes in the diameter or thickness of the crystal. The optimization of the technological process consists in creating the possibility of controlling the crystallization front shape with minimal temperature gradients in the crystals, but at sufficiently high growth rates. For purposeful control of crystal growth there is a need in reliable data on the interfaced processes of heat exchange between a crystal, melt and environment [1-5]. Controlling the thermal growth conditions of crystals is a rather difficult task, since as the length of the crystal increases, the heat transfer conditions from its surface to the environment change. From the point of view of setting the boundary value problems of numerical simulation, this is equivalent to a continuous change in the geometry of the computational domain. Accordingly, the conditions of heat transfer from the crystal in the regimes of complex conjugate heat exchange throughout the growing vessel change. The complex of unresolved problems of heat exchange largely determines the difficulties of creating well-controlled technologies for growing single crystals. To solve the problem of the dependence of crystallographic, optical and electrophysical characteristics on crystal growth conditions, it is necessary to be able to predict the temperature fields and the temperature gradients inside the crystals [1-5].

The parametric studies of radiation-convective heat transfer from flat and cylindrical crystals were carried out numerically, in idealized formulations, the relative role of the thermal conductivity, convective and radiation heat transfer mechanism was determined. Simulation of convective heat transfer was carried out on the basis of a system of equations of natural convection in Boussinesq approximation in term of temperature, vortex, and stream function. The calculations were carried out by the finite element method [6]. The method of consistent results [6] was used to calculate the values of the vortex and velocity, which allowed obtaining the values of partial derivatives of an arbitrary...
finite element solution with high accuracy. Radiation fluxes were calculated on the basis of the zonal method [7]. The relative role and cumulative effect of various heat exchange mechanisms (thermal conductivity, convection and radiation) on the temperature fields in crystals depending on the characteristic temperature difference is studied.

2. Problem definition

For growing vessel used in the growth of single crystals by the Czochralski method and floating zone method (FZ), axial symmetry is characteristic (figure 1a, 1b). Taking into account the properties of axisymmetry, the calculations are carried out in two-dimensional regions in cylindrical coordinates. This has significantly reduced the cost of computing.

For the geometry used for growing monocrystalline tapes (figure 1c), numerical simulation is carried out in two-dimensional formulation in Cartesian coordinates. Numerical simulation is carried out by the finite element method on the nonuniform grid of triangular finite elements with linear functions. For Czochralski method a mesh with 100х500 nodes was used. A nonuniform grid with 16948 nodes was used for FZ. To simulate thermogravitational convection, we used the dimensionless system of Navier-Stokes equations, energy and continuity in the Boussinesq approximation, written in variable vortex, stream function and temperature, in a cylindrical coordinates:

\[
\begin{align*}
-\frac{1}{Pr} \left( \frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial z^2} \right) + u \frac{\partial T}{\partial r} + v \frac{\partial T}{\partial z} &= 0 \\
- \left( \frac{\partial^2 \omega}{\partial r^2} + \frac{1}{r} \frac{\partial \omega}{\partial r} + \frac{\partial^2 \omega}{\partial z^2} \right) + u \frac{\partial \omega}{\partial r} + v \frac{\partial \omega}{\partial z} + \frac{\omega}{r^2} - u \frac{\omega}{r} &= Gr \frac{\partial T}{\partial r} \\
- \left( \frac{\partial^2 \psi}{\partial r^2} + \frac{1}{r} \frac{\partial \psi}{\partial r} + \frac{\partial^2 \psi}{\partial z^2} \right) + \frac{\psi}{r^2} &= -\omega
\end{align*}
\]

Figure 1. Computational domain for: a - Czochralski method, b - floating zone method, c – Stepanov’s method.
where $T$, $\omega$ and $\psi$ are the temperature, vorticity and stream function, respectively, and $u$ and $v$ are the radial/axial and horizontal/vertical components of velocity, respectively.

In dimensionless equations $Gr = \left( \frac{\beta g \Delta T}{\nu^2} \right) \times R_k^3$ - the Grashof number, by definition, is the ratio of buoyancy forces (Archimedes forces) to the forces of viscous friction, and given the geometry and parameters of the gas, it can be treated as a dimensionless temperature difference. The Prandtl number $Pr = \frac{\nu}{\lambda}$, $\lambda = \frac{\lambda}{\rho C_P}$ is the thermal diffusivity, $g$ is the acceleration of gravity, $\beta$ the coefficient of volume expansion of the gas, $\rho$ is the density, and $C_P$ is the specific heat at constant pressure.

When reducing the equations to the dimensionless form, the radius of the crystal $R_S$ is used as the geometric scale for the modeling of heat transfer in the Czochralski and floating zone methods. When modeling the ribbon pulling from a melt by Stepanov’s method, the distance from the center of the growth chamber to the cold wall of growth chamber is taken as the geometrical scale. As a temperature scale, the temperature difference between the crystallization front and the walls of the growth vessel was chosen. The velocity field is scaled by $v/R_S$. Scale for radiative fluxes is $R_S/\lambda_{gas}\Delta T$.

For Stepanov’s method (figure 1c), the grid with 17557 nodes was applied. For the simulation of thermo-gravitational convection we used the dimensionless Navier-Stokes equations in the Boussinesq approximation, written in the variables vortex, stream function and temperature. In Cartesian coordinates:

$$
-\frac{1}{Pr} \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right) + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} = 0
$$

$$
- \left( \frac{\partial^2 \omega}{\partial x^2} + \frac{\partial^2 \omega}{\partial y^2} \right) + u \frac{\partial \omega}{\partial x} + v \frac{\partial \omega}{\partial y} = Gr \frac{\partial T}{\partial x}
$$

$$
\left( \frac{\partial^2 \psi}{\partial x^2} + \frac{\partial^2 \psi}{\partial y^2} \right) = -\omega
$$

The calculation of radiation fluxes was carried out under the following assumptions: the calculation area is limited to a closed system of surfaces; all surfaces of the system are gray, diffuse-emitting and diffuse-reflective; surfaces are divided into zones within which the radiation properties and temperature can be considered constant; and the medium filling the growth vessel are diathermic.

The maximum temperature in the system is set at the crystallization front. The walls of the growth chamber are maintained at the minimum temperature in the system. They set the condition of non-flow and adhesion. For simulated variants of the methods of Czochralski and Stepanov it is considered that the surface of the melt is closed from the surrounding medium by a screen. On the surface of the screen, the conditions for thermal insulation, non-flow and adherence are set. The conditions of non-flow, adherence and the condition of ideal contact taking into account radiation fluxes are set on the crystal generatrix:

$$
-\lambda_r \frac{\partial T}{\partial n} = -\lambda_{gas} \frac{\partial T}{\partial n} + Q
$$

$$
T|_+ = T|_- .
$$

3. Experimental results

The essence of the FZ method is that the lower end of the polycrystalline silicon rod is melted by an inductor. A stream of melt flows down to the upper end of the single crystal. At the end of the single crystal, the central part remains molten during crystal growth from the bottom up. The crystallization front of the melt has a parabolic shape concave into a single crystal. The melt is retained by crystallization of the peripheral part of the crystal generatrix. Monocrystal generators are cooled in the regime of radiation-convective heat transfer into the surrounding gas environment and to the cold walls of the growth vessel.
In this paper, calculations are carried out without taking into account the convective flow in the melt, which has a thermal gravitational-capillary character and is subject to the influence of inductor radiation. The main attention is paid to the thermal history of the crystal, which affects the quality of the resulting single crystal, the distribution of impurities, dislocations and thermal stresses in the crystal.

The results of calculations for FZ are presented in figure 2. Since the process of FZ is high-temperature and is carried out in an inert gas atmosphere with high pressure, the contributions of radiation and convective heat transfer mechanisms can be equivalent. The position of the molten zone affects the development of convective flow of gas. Depending on the length of the crystal and polycrystalline, the flow regimes change significantly. The local structure of the gas flow in the transition zone from polycrystalline to melt and single crystal varies the most. In the area below the melt zone, the longitudinal temperature distribution creates an accelerating convective flow, in the upper part, on the contrary, the heated gas flows into the cold wall of the polycrystalline, warming it up. Accordingly, the local temperature fields and heat fluxes change. Temperature gradients vary greatly in the single crystal depending on its length. When accounting for the radiative heat transfer, the local heat fluxes from the crystal generatrix increase and the axial and radial temperature gradients are significantly rise. In general, the single crystal is cooled more effectively.

For the Czochralski method geometry appropriate to real technology, in [4] calculations in the range of temperature changes equivalent to the range of the Grashof number $1000 \leq \text{Gr} \leq 16000$ are carried out. $\text{Gr} = 16000$ corresponds to a temperature difference $\Delta T = 1330$ K.

Accounting of radiation fluxes significantly changes the temperature distribution on the surface and inside the crystal, the local heat fluxes from the crystal surface increase noticeably. Cooling efficiency increases throughout the crystal volume, as can be seen from the closeness of the isotherms at the base of the crystal (figure 3a). The temperature on the crystal generatrix decreases, and the temperature difference between the walls of the growth vessel and the surface of the crystal decreases. As a result,

![Figure 2](image_url)

**Figure 2.** Fields of isotherms and isolines of the stream function in the method of FZ with $\text{Gr} = 1008519$, $\text{Pr} = 0.63$, $\Delta T = 1330$ K, $T_{\text{max}} = 1680$ K, a - isolines of stream function (left) and isotherms in convective regime, b - isolines of stream function (left) and isotherms in radiative-convective regime, c - isotherms in the convective (left) and radiative-convective (right), d - isotherms in the conductive (left) and radiative-convective (right) regime.
at a given characteristic $\Delta T$ (or $Gr$), the intensity of convective flows decreases slightly, but the spatial form of convective flows remains (figure 3b). The axial temperature gradients increase significantly (figure 4a), and the shape of their radial distributions at the crystallization front changes. Radial temperature gradients increase (figure 4b). The relative role in the cooling of the heat sink crystal is changed by means of a conductive mechanism through the seed crystal and the rod and heat transfer from the side surface of the crystal. For the high thermal conductivity of the crystal, the heat flux through the side surface is about 0.6% of the total heat flux (for the crystal height equal to 1) up to 2.5% (for the crystal height equal to 4) for the convective regime without radiation. Taking into account the radiation greatly increases the heat fluxes, up to 22% and 65% respectively.

Calculations of radiation-convective heat transfer from monocrystalline ribbon to cold walls of the case are carried out in the field of such simplified technological geometry of the growth node of Stepanov's method taking into account and neglecting the dependence of thermal conductivity of sapphire on temperature. The simulation was carried out with the number of Grashof $Gr = 25404$ ($\Delta T = 1970$ K) and in the range of monocrystalline ribbon lengths $1 \leq H/L \leq 5$.

In a backlash between a ribbon and cold walls of the case the circulating flow of gas is established (figure 5a, 5b). The gas is heated at the base of the tape (crystallization front), rises up the crystal and reaches the cold cover of the case. Then the gas flow turns around, reaches the cold walls of the housing, cools and falls to the screen. After that, the cold gas stream unfolds and runs on the heated base of the single crystal tape. As a result, the cooling efficiency of the belt increases significantly.

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**Figure 3.** Isotherms (a) and isolines of the current function (b) in convective (right) and radiation-convective mode (left). Additional isotherms 0.975 and 0.9875 have been added to the interval between the dimensionless isotherms 1 and 0.95 strokes.

**Figure 4.** Axial (a) and radial (b) temperature gradients in convective (1-3) and radiation-convective (4-6) regimes at crystal length 4 at levels (b): 1 – 4-z = 0; 2, 5 – 1; 3, 6 – 1.5, and sections (g): 1, 4-r = 1; 2, 5 – 0.8; 3, 6 – 0.5.
Figure 5. Isolines of stream function (left) and isotherms (right) in the convective (a) and radiative-convective (b) regime. Isotherms in convective (left) and radiation-convective (right) regime (c).

near the crystallization front, and longitudinal and transverse temperature gradients increase sharply. Taking into account the radiation mechanism of heat exchange leads to a more efficient cooling of the crystal.

4. Conclusion
Conjugate radiation-convective heat exchange of the crystal with the environment in a system geometrically similar to the intermediate stage of the technological process of growing single crystals by methods of Czochralski, Stepanov and FZ has been investigated. With fixed geometry of the computational domain, the evolution of the flow structure and conjugate convective heat transfer in the modes of thermal conductivity, natural convection and radiation-convective heat exchange with an increase in temperature difference have been investigated. The effect of heat transfer regime on the temperature fields in the crystal has been studied. The relative roles of thermal conductivity, radiation heat transfer and convective heat transfer in the gas phase have been investigated. It is shown that in the investigated range of temperature changes the role of convective heat transfer remains essential for all considered methods of growing single crystals. Radiation heat transfer lowers the temperature of the crystal surface and reduces the temperature difference between the crystal generatrix and the walls of the growth chamber. As a result, the intensity of convective motion of the gas decreases. The influence of flow characteristics on the agreed temperature fields in the gas and in the crystal has been investigated. With small numbers of Grashof, the effect of convection on the temperature distribution in the gas is relatively small.

As the length of the monocrystalline ribbon increases, the temperature fields and temperature gradients change significantly in Stepanov's method, followed by thermal stress fields in the volume of the resulting crystal. A significant influence on the temperature field has convective heat transfer
mechanism. With the growth of the monocristalline ribbon length, the spatial form of convective flows changes. Zones of separation of a boundary layer which considerably change local regularities of heat transfer can be formed. At the same time, starting from some tape length, in the lower region of the growth chamber, the spatial form and intensity of convective flows are established. In other words, the formed flow parameters do not change with increasing tape length. It is shown that the transverse distribution of the longitudinal temperature gradient in the monocristalline ribbon near the crystallization front is expressed nonuniformly.

In FZ, the position of the molten zone significantly affects the development of the convective flow in the growth chamber. Depending on the length of the crystal and polycrystalline, the flow regimes change significantly. The local structure of the gas flow in the transition zone from polycrystalline to melt and single crystal varies the most. In the area below the melt zone, the longitudinal temperature distribution creates an accelerating convective flow, in the upper part, on the contrary, the heated gas flows into the cold wall of the polycrystalline, warming it up. Accordingly, the local temperature fields and heat fluxes change. Temperature gradients vary greatly in the single crystal depending on its length. When accounting for the radiative heat transfer, the local heat fluxes from the crystal generatrix increase and the axial and radial temperature gradients significantly rise.

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