Numerical studies of the effect of the temperature drop in the crucible - melt - cooled disk system on the shapes of crystallization fronts

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Abstract. The crystallization process on a cooled disk located on the free surface of a water layer is studied numerically. The influence of thermal gravitational-capillary and mixed convection on the shape of the crystallization front 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 the problem by the finite element method using an adaptive triangular grid and taking into account the latent heat of crystallization and the inverse dependence of density on temperature.

1. Introduction

When growing single crystals by the Czochralski and Kyropoulos methods, crystallization methods are used on a seed crystal touching the free surface of the melt without contact with the crucible [1, 2]. As for all technological systems containing nonisothermal liquids or melts and located in a gravity field, thermogravitational convection occurs in the thermal units of growth plants [1-5]. In the classical versions of the Czochralski method with a fixed crucible, the rotation of the crystal is used to control the technological process in addition to heating the walls of the crucible, cooling the seed and the crystal [1-3]. A radial temperature gradient arises on the free surface of the melt between the cold edge of the crystal and the heated wall of the crucible. The temperature dependence of the surface tension of the melt initiates the development of thermocapillary convection [4, 5]. Thus, to control the crystallization process and the shape of the crystallization front (CF), it is necessary to select the modes of conjugate convective heat transfer by controlling the crystal rotation and temperature drops. To obtain high-quality crystals, the initial stage of the crystal growth process and reaching a given diameter is critical. The overwhelming majority of melts have a normal dependence of density on temperature. But melts of some crystals, important for the creation of modern thermal imaging technology have an inverse dependence of density on temperature. Water is a convenient model liquid-simulator of such melts in the experimental and numerical study of the features of their crystallization [6, 7]. Numerical simulation of the initial stage of crystallization of water with an inverse dependence of density on temperature on a cooled disk has been performed.
2. Problem statement

In studies of convective heat transfer during crystal growth, until now there are few works in which conjugate convective heat transfer is studied in cases when a complex of buoyancy forces, thermocapillary effect (TCE), and rotation effects are acting. Due to the nonlinearity of the interaction of various flow generation mechanisms, many unresolved questions remain. The effect of the inverse dependence of the density of melts on temperature on the features of hydrodynamics and heat transfer in crystallization modes has been poorly studied. Such properties are possessed, for example, by mercury cadmium tellurides [6, 7], whose crystals are widely used in modern technology. The initial stage of the process of growing a seed crystal to a given diameter is critical. Numerical studies of unsteady conjugate convective heat transfer on the crystallization process and the forms of CFs make it possible to obtain practically exhaustive data on the instantaneous temperature fields in the liquid and solid phases. Numerical simulation of the initial stage of crystal growth from a melt with the inverse temperature dependence of density has been performed. Water is a convenient model liquid that simulates such melts for experimental and numerical studies of the features of their crystallization.

The diagram of the computational domain in the numerical formulation of the problem (Figure 1, the right side of the axisymmetric system is shown) consists of a cylindrical crucible ($\Omega_3$) with a given wall and bottom thickness, melt, and crystal, at a temperature on the disk surface $S_{10}$ below the crystallization temperature. A stationary or rotating disk is instantly cooled at time $t \geq 0$ to $T_D = -5 ^\circ C$. The boundary of the disk $S_{10}$ is located at the level of the free surface of the liquid ($S_3$). On the outer side of the sidewall of the crucible ($S_7$), the initial temperature of the system is maintained at $T^* = +10 ^\circ C$. The inner radius and height of the crucible are 5 cm. Wall thickness is 0.5 cm. The disk radius is 2 cm. The calculations were carried out for a stationary disk and at a disk rotation speed $\omega_D = 2$, 5, and 10 rpm. The finite element method [8] in a nonstationary axisymmetric conjugate formulation of problems was used to solve dimensionless systems of equations for thermal gravitational-capillary and mixed convection, and dimensionless equations of heat conduction in the crystal and the crucible walls in terms of temperature, a vortex of velocity, stream function, and the azimuthal component of velocity. The system of equations and boundary conditions are similar to the formulation of the problem for the silicon-graphite crucible system [5]. Simulation of the crystallization process was carried out for water in a plexiglass crucible taking into account the inverse dependence of water density on temperature [9]. Accounting for the heat of crystallization during crystallization or melting occurs through an internal source of heat release or absorption. The zones of these sources are redefined in the iterative process of the conjugate solution of the system of equations within the current time layer. After establishing the solution for the current time layer, the triangular grid is rebuilt, where the crystallization front is formed by the edges of the triangular elements. The grid thickens towards all boundaries, including the crystal-melt interface. The number of mesh nodes reached up to 27,984, triangular elements – up to 54,949. At the interfaces between the "crucible-melt", "crucible-crystal", "melt-crystal" media, the conditions of ideal thermal contact, i.e., the continuity of temperature and heat flux, are satisfied. The no-slip conditions are satisfied on rigid surfaces. The upper horizontal boundary of the computational domain $S_{11}$ and the lower horizontal boundary $S_6$ are adiabatic. At the initial moment, the temperature in the entire system is constant and there is no convective flow.

![Figure 1. Scheme of the computational domain.](image-url)
3. Results and discussion
In the modes of non-stationary thermal gravitational-capillary convection, the process of water crystallization on the surface of a monotonically cooled disk after reaching the crystallization temperature is investigated. Thus, the effect of unsteady natural convection, excited by the combined effect of buoyancy and TCE forces, on the hydrodynamics of the melt, heat transfer, and the shape of the CF 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, the forced flow occurs when the cooled disk rotates uniformly.

Figure 2a shows the fields of isotherms and isolines of the stream function, established by the time \( t = 2275 \) s in the mode of thermal gravitational-capillary convection. Figures 2 b-d show the dependences of the spatial form of the flow and the corresponding fields of isotherms on the disc rotation speed at the same fixed moment. It is clearly seen here that from the regime of thermal gravitational-capillary convection to the regime of developed mixed convection along the free surface there is a flow of a thermocapillary nature. The flow of the liquid heated on the crucible walls reaches the edge of the crystallized water even with a developed forced flow under the rotating CF (Figures 2 c, d). Under similar conditions in a liquid with \( Pr = 16 \), a centrifugal flow from under the CF displaces the free-convective flow flowing along the free surface from the crystal edge [3, 4]. Figure 3 shows the radial temperature distributions along the free surface of the water layer. The temperature gradient along the free surface of the water layer, which has abnormally high surface tension and is temperature-dependent, causes a significant contribution of the TCE to the generation of a heated liquid flow to the edge of the CF. Figure 3a shows the temperature distribution from the crucible wall to the crystal edge and in the solid. Figure 3b shows, on a large scale, the temperature distribution at the edge of the crystal (section from \( T \approx 10 \) °C to \( T = 0 \) °C) and in a solid (section with \( 0 \) °C \( \leq T \approx -5 \) °C). These data (Figures 2, 3) can be used to determine the temperature gradients in the crystal.

The velocities of the heated liquid flowing along the free surface onto the edge of the CF are shown in Figure 4a. According to the data in Figures 3, 4a, it can be seen that with an increase in the CF rotation rate, the temperature on the free surface decreases due to the removal of the cooled liquid from under the CF. But the longitudinal temperature gradient decreases insignificantly. The formation and intensification with an increase in the rate of rotation of the CF of the flow, which flows onto the CF from below, has a more significant effect on the shape of the CF. The radial distributions of the axial velocity component in Figure 4b allow understanding of spatial structure and intensity of the fluxes flowing onto the CF. The isotherm fields in Figure 2 and the profiles of the velocity components in Figures 3-5 give a clear idea of the reasons for the dependence of the CF shapes on the rotation rate for a given characteristic temperature drop in the system.

The rotation of the disk and the CF after the appearance of a layer of crystallized matter, in addition to creating an ascending fluid flow in the axial region, affects the flow conditions around the crucible walls. The upward flow of the heated liquid on the sidewall of the crucible acquires an azimuthal velocity component in the outer part of the boundary layer (Figure 5). The conditions and intensity of convective heat transfer on the walls and at the bottom of the crucible change. The conditions for heat transfer through the core of the liquid layer from the heated walls of the crucible to the CF change. This is clearly seen from the changes in the isotherm fields during the transition from a stationary CF rotating at a low speed (Figures 2 a, b) to a pronounced gravitational-centrifugal regime at \( 5 \) rpm \( \leq \omega_D \) \( \leq 10 \) rpm (Figures 2 c, d). At \( \omega_D = 10 \) rpm of a forced nature, the flow already flows around a part of the heated wall of the crucible, and therefore the convective heat flux to the CF increases. The radial size of the heated liquid flow incident on the CF increased sharply. As a result, the volume decreases while the thickness of the solidified substance layer is aligned in the radial direction. The dependences of the CF shapes on the characteristic temperature drops and at each of them on the rotation rate are shown in Figure 6. With an increase in the characteristic temperature drop, the growth of a crystal with an almost flat crystallization front is possible, but with an increase in the crystal rotation rate. This can be seen from the data in Figures 6 c, d.
Figure 2. Fields of isotherms (left) and isolines of the stream function (right) at time \( t = 2275 \, s \), depending on the disc rotation speed: a – \( \omega_T \, (rpm) = 0 \); b – 2; c – 5; d – 10. \( T_D = -5 \, ^\circ C \).
Figure 3. Temperature distribution at time $t = 2275$ s in section $z = 5.5$ cm at disk rotation speeds: 1 – $\omega_D$ (rpm) = 0; 2 – 2; 3 – 5; 4 – 10. $T_D = -5$ °C. (a – normal scale, b – large scale).

Figure 4. Dependences on the disk rotation speed of the radial distributions of the horizontal velocity component (a) at the free boundary $z = 5.5$ cm and the vertical velocity component (b in the section $z = 4$ cm at the time $t = 2275$ s: 1 – $\omega_D$ (rpm) = 0; 2 – 2; 3 – 5; 4 – 10.

Figure 5. Profiles of the azimuthal velocity component at the time $t = 2275$ s in sections $z = 5.5$ cm (a) and $z = 4$ cm (b) at disk rotation speeds: 1 – $\omega_D$ (rpm) = 0; 2 – 2; 3 – 5; 4 – 10. $T_D = -5$ °C.
Figure 6. Forms of crystallization fronts at time $t = 2131$ s at temperatures on the disks: $1 – T_D (\degree C) = -2$; $2 – -5$; $3 – -10$; at angular speeds $\omega_D (rpm)$: $a – \omega_D = 0$; $b – \omega_D = 2$; $c – \omega_D = 5$; $d – \omega_D = 10$.

4. Conclusion
The initial stages of the unsteady process of water crystallization during sudden cooling of the disk to $-2$, $-5$, $-10$ °C are numerically investigated. The initial temperature of the system is 10 °C. The unsteady modes of thermal gravitational-capillary and gravitational-centrifugal convection were studied (at disk rotation speeds equal to 2, 5, and 10 rpm). In the thermal gravitational-capillary and mixed gravitational-centrifugal modes, the presence of the thermocapillary effect and the inverse dependence of density on temperature affects heat transfer and the shape of the crystallization front. The results obtained can be useful for growing single crystals from melts, for example, the cadmium-mercury-tellurium eutectic.

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