Numerical simulation of the crystallization front propagation during the bottom cooling of a flat layer of water

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Abstract. The process of crystallization of water after a sudden cooling of the bottom of a rectangular cavity is investigated by numerical simulation of conjugate convective heat exchange using the finite element method. Detailed information has been obtained on hydrodynamics, temperature fields, and the forms of the crystallization front. The crystallization of water is studied by taking into account the dependence of the density on the temperature and heat of the phase transition. Numerical studies are carried out using a proprietary software package. Adaptive triangular grids are used to track the position of the crystallization front at each time step.

1. Introduction
The cultivation of single crystals is the next stage after the synthesis of a new material. Even small-sized single crystals are required to obtain samples to determine their basic properties and suitability for laser technology development, nonlinear optics or for using other optoelectronic properties of a new crystal [1, 2]. The next step consists of increasing the size of a grown crystal, scaling and bringing the technology to the stage of industrial production. At each stage it is important to determine the optimal technological growth regimes of the single crystals. An understanding of local laws of heat and mass transfer that depend on the spatial form of convective flows [2-6] plays a critical role in determining the optimal parameters. The shapes of the crystallization fronts at different stages of crystal growth depend on the local features of the conjugate convective heat exchange. Due to the fact that growth bands and other imperfections are unambiguously associated with intermediate forms of crystallization fronts, an understanding of the characteristics of heat transfer is necessary to create controlled technological process [2, 3]. Water is considered as a liquid-simulator of melts with inverse density dependences on temperature. An example of such melts is the cadmium-mercury-tellurium alloy and some pure substances [7, 8]. To date cadmium-mercury tellurides (CRT) occupy a leading position among the materials used in the creation of devices for recording infrared radiation, especially thermal imagers [8]. Amongst the many problems needed to be solved to create the technology of growing high-quality and sufficiently large-sized single crystals of CRT in the mentioned papers and many others, the features of hydrodynamics and heat transfer in the presence of inverse density dependence on temperature are not examined. The solution of this problem requires deep understanding of the features of convective heat transfer in the presence of an inverse dependence of density on temperature as well as with various combinations of thermal conductivities of melts and crucible materials. The research in this paper focuses on such problems. One of the most commonly used methods for producing single crystals is the vertical Bridgman method [1, 2, 4]. In this method,
the melt is superheated relatively to the crystallization temperature of the substance and then lowered from the upper part of the furnace at a given speed into the cold zone at the bottom of the furnace. A crystal begins to form on the cooled bottom. The containers in which the melt is located can have various shapes of the bottom part, including the flat ones [5, 6]. To obtain crystals for solar energy, rectangular crucible containers are used [5, 6]. It is almost impossible to obtain detailed information on local heat exchange process at different stages of real crystal growth. Numerical simulation allows us to obtain in-depth information on velocity and temperature fields as well as crystallization fronts. The results have been obtained using the software package developed by the authors of this paper.

The system of equations of unsteady free convection is solved with consideration of both the dependence of water density on temperature and the heat of phase transition. In a solidified substance the equations of unsteady heat conduction are used. Solutions are obtained in conjugate heat exchange regimes under the continuity condition for the temperature fields and heat fluxes.

2. Statement of the problem

The computational domain is shown in Figure 1. The size of the computational domain is set to 105 × 105 mm². Convective heat transfer in the melt is described by the system of equations of thermogravitational convection in the Boussinesq approximation written in dimensionless variables of temperature, vector potential of the velocity field, velocity vortex in the Cartesian coordinate system:

\[
\begin{align*}
\left( \frac{\rho(T)}{\rho_1} \right) \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} \right) &= \frac{\lambda_1}{\rho_1 c_v V_1} \left( \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial x^2} \right), \\
\left( \frac{\rho(T)}{\rho_1} \right) \left( \frac{\partial \omega}{\partial t} + u \frac{\partial \omega}{\partial x} + v \frac{\partial \omega}{\partial y} \right) &= \left( \frac{\partial^2 \omega}{\partial y^2} + \frac{\partial^2 \omega}{\partial x^2} \right) + g \cdot \beta(T) \cdot \frac{H^3(H_{\max} - T_{\min})}{V_1^2} \frac{\partial T}{\partial x}, \\
\frac{\partial^2 \psi}{\partial y^2} + \frac{\partial^2 \psi}{\partial x^2} &= -\omega,
\end{align*}
\]

where \( \beta \) is the coefficient of volumetric thermal expansion coefficient, \( g \) is the acceleration of gravity. The velocity vortex and the stream function are related to the velocity components as follows:

\[
\omega = \frac{\partial v}{\partial x} - \frac{\partial u}{\partial y}, \quad u = \frac{\partial \psi}{\partial y}, \quad v = -\frac{\partial \psi}{\partial x}.
\]

The following boundary conditions are given:

1. At the “wall–melt” and “wall–crystal” interfaces perfect thermal contact is assumed, i.e. at the specified boundaries the temperature and heat flux are continuous:

\[
\lambda_j \frac{\partial T}{\partial n} \bigg|_{\Gamma_{i+}} = \lambda_{2,j} \frac{\partial T}{\partial n} \bigg|_{\Gamma_{i-}}, \quad T \bigg|_{\Gamma_{i+}} = T \bigg|_{\Gamma_{i-}} \quad (i = 2, 4, 5, \text{see figure } 1).
\]

2. At the “melt – crystal” interface the Stefan condition is used:

\[
\lambda_j \frac{\partial T}{\partial n} \bigg|_{\Gamma_{i+}} = \lambda_2 \frac{\partial T}{\partial n} \bigg|_{\Gamma_{i-}} + \rho LV,
\]

where \( V \) is the normal velocity of a point lying on the phase boundary, \( L \) is the latent heat of the phase transition.

3. At the non-deformable boundaries of the melt region, no-flow conditions are given:

\[
\psi \bigg|_{\Gamma_{i+}} = 0 \quad (i = 1, 2, 3, 4).
\]

Similar conditions are set at the “crystal–melt” boundary:

\[
\psi \bigg|_{\Gamma_{5}} = 0.
\]

4. On the internal hard surfaces of the walls in the liquid and on the “crystal–melt” boundary, the adhesion conditions are used, as a result the condition for the velocity vortex is written as:
5. The temperature at horizontal boundaries in the fluid \((\Gamma_1, \Gamma_3)\) at the initial time instant is set at constant values: \(T|_{\Gamma_1} = -10^\circ C\), \(T|_{\Gamma_3} = +10^\circ C\).

\[
\omega_{i,1} = -\frac{\partial}{\partial x} \frac{\partial \psi}{\partial x} - \frac{\partial}{\partial y} \frac{\partial \psi}{\partial y}, \quad i = 1, 2, 3, 4, 5.
\]

6. The outer boundaries of the lateral plexiglass walls are always set as adiabatic. The results given below in this paper have been obtained using adiabatic inner surfaces:

\[
\frac{\partial T}{\partial x}|_{W_{2,5}} = 0, \quad \frac{\partial T}{\partial y}|_{\psi|_{1,3,4,6}} = 0.
\]

7. At the initial moment of time the temperature in the entire system is set at a constant value of the temperature on the hot horizontal wall \(+10^\circ C\), no initial convective currents occur:

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

In the presented problem the latent heat of the phase transition is considered. Therefore the energy equation in the crystallization zone is modified as follows:

\[
\frac{\rho_2}{\rho_1} \left( \frac{c_2}{c_1} + \frac{L}{c_1 \Delta T} \right) \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} \right) = \frac{\lambda_2}{\rho_f c_{pf} \nu_f} \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right).
\]

At each time step the boundaries of the crystallization zone are decided by the position of the “crystal–melt” interface, the new position of the front is determined during the iterative process of solving equations. The finite element method discretisation is applied to the system of equations. Linear functions on triangles are chosen as basis functions. The calculations are performed on an adaptive grid that consists of 15638 nodes and 30751 elements. The triangular grid tracks the position
of the crystallization front at each time step and is refined in its vicinity, as well as next to all the boundaries of the computational domain to a varying degree. Figure 2 shows a fragment of the grid. The size of the presented grid fragment is 0.01 × 0.17 in dimensionless form (corresponding to 21.105 mm × 17.85 mm).

3. Results and discussion

Figures 3 and 4 show the temporal evolution of the temperature field and the shape of the crystallization front. The isotherm of +4°C is indicated. The critical value of the Rayleigh number is estimated \( Ra_c = 680.6 \), according to the data in the time range of \( 18.45 \text{ min} \leq t \leq 23.06 \text{ min} \). After violation of the stability of mechanical equilibrium in the water layer above the upper ice boundary, the Rayleigh-Benarow cellular convection develops. Over time, the intensity of the convective flow increases and the curvature of the crystallization front appears. The concave sections correspond to the downward fluid flow at a higher temperature. It shows a distinctive feature of the development of convection in water in the temperature range from 0°C to +4°C. The liquid has a lower temperature in the ascending flow. These flows correspond to the convex sections of the crystallization front. The isoline fields of the stream function in Figure 5 show a high degree of spatial correlation of the temperature fields in Figures 3 and 4 with the spatial flow pattern. When the spatial shape of the flow is being restructured, as shown in Figures 3 and 4, the local curvature of the crystallization front follows.

With an increase in the thickness of the ice layer and propagation of the isotherm of +4°C into the upper water layers, the flow loses its initial highly coherent spatial form. With the growth of supercriticality the instability develops due to the competition of cells in a layer limited in length. It is caused by the increase in the thickness of the water layer with unstable density stratification as growth of supercriticality always leads to an increase in the wavelength of a cellular flow. In an essentially steady state at \( t = 160.59 \text{ min} \) only one asymmetric convective cell remains. In this state, the position of the crystallization front does not change.
Figure 4. Isotherm fields in the near-bottom layer at different moments in time.

Figure 5. Stream function isoline fields in the near-bottom layer at different moments in time.

Figure 6 (left) shows the temperature distribution along the height of the entire computational domain in sections along the horizontal coordinate corresponding to the positions of the ascending and descending flows in a separate cell, red-lined section corresponds to the center of rotation of the fluid. On the right figure 6 shows the temperature distribution in the same sections in an enlarged scale within the height of the cell. It is worth to note the almost neutral stratification of the water layer in its upper part. In separate shafts of cells, temperature distributions are typical for finite-amplitude cellular Rayleigh-Benard convection. In the central part reverse temperature gradient is observed.

Figure 7 shows the distribution of the vertical speed component at different time moments, including the moments shown in Figures 5 and 6. The sectional value of the vertical coordinate is chosen at the level of the center of rotation in the shaft of cells in Figure 5. The amplitudes of the vertical component of velocity increase with supercriticality. Therefore, descending convective heat fluxes from the upper layers of the liquid to the crystallization front increase. This leads to an increasing heterogeneity of the ice layer thickness as seen in Figure 8 that shows the evolution of the
ice layer thickness and the shape of the crystallization front with time after a sudden cooling of the lower boundary of the layer to -10°C.

Figure 6. Temperature distribution at different moments in time (non-scaled on the left, scaled on the right).

Figure 7. Distribution of the vertical speed component in section y at times t (min.):
1 – z = 0.23; t = 25.62; 2 – 0.29; 32.8; 3 – 0.35; 44.63.

Figure 8. Evolution of the ice layer thickness and the shape of the crystallization front with timeranging from t = 307.44 s to t = 6148.8 s at intervals of 307.44 s.
At the initial stage in the thermal conductivity mode, the crystallization front is flat-shaped. After the onset of cellular convection, the front curvature has regular nature. Upon transition to a substantially unsteady flow regime and the formation of one asymmetric cell, the front shape becomes anomalously concave.

Conclusion
A finite element method is used to carry out numerical modeling of conjugate convective heat transfer in a rectangular cavity, the bottom being suddenly cooled to a temperature below the crystallization temperature of water. Algorithms for constructing adaptive grids have been implemented. The process of crystallization of water – simulator of melts with inverse dependence of density on temperature – is studied. The shape of the crystallization front and the growth rate of the mass of the solidified substance depend on the processes of conjugate convective heat exchange at the interface. Obtained results can be used in analyzing the processes of crystal growth in methods of directional crystallization of substances, including substances with the inverse dependence of the melt density on temperature (e.g. cadmium-mercury-tellurium, gallium, bismuth crystals).

Acknowledgments
This research is carried out as a part of the project III.18.2.5, number of state registration AAAA-A17-1170228500213, and under RFBR project number 18-38-00790 мол_а.

References
[1] Wilke K-T 1977 Growing crystals (Leningrad, Nedra, Leningrad branch)
[2] Mokhtar Ben Sassi, Slim Kaddeche, Marcello Lappa, Séverine Millet, Daniel Henry, Hamda Ben Hadid 2017 The Journal of Crystal Growth 458 154–65
[3] Anfimov I M, Berdnikov V S, Vygovskaya Ye A, et al. 2007 Izv. Universities. Materials electron. technology 4 40–4
[4] Antonov P V, Berdnikov V S 2012 Applied Mechanics and Technical Physics 53 (6) 65–77
[5] Meier D, Lukin G, Thieme N, Bönisch P, Dadzis K, Büttner L, Pätzold O, Czarske J, Stelter M 2017 Journal of Crystal Growth. 461 30–7
[6] Nepomnyashchikh A I, Presnyakov R V, Antonov P V, Berdnikov V S 2015 News of the Universities. Applied chemistry and biotechnology 1 (12) 11–7
[7] Glazov V M, Pavlova L M, Stankus S V, Khairulin R A 1997 Reports of the Academy of Sciences 354 (2) 207–10
[8] Ponomarenko I P 2003 UFN 173 (6) 649–65
[9] Vargaftik N B 1972 Spravochnik po teplofizicheskim svoistvam gazov i zhidkostei (Handbook on Thermophysical Properties of Gases and Liquids) (M.: Nauka)