Heat field rotation method of crystal growth: numerical simulations

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Abstract

In that paper we present results of numerical simulation of convective flows which exist in BSO (Bi₁₂SiO₂₀) and Si melts while growing crystals in Czochralski configuration in a rotating heat field of first order axial symmetry (rotL₁). The numerical simulation is based on the solution of non-stationary three-dimensional Navier-Stokes equations and heat-transfer equations in Boussinesq approximation. The results showed that heat field rotation makes possible an efficient mixing of molten material without any mechanical tools. Trajectories of closed toroidal flows produced by contact-free control over heat-mass transfer process and having the azimuthal component of velocity vector have been simulated in the melts.

1 Introduction

The recent publications and patents [1-5] have shown that Heat Field Rotation Method (HFRM) is a principally new and efficient method for control over heat-mass-transfer processes in crystal growth. The method implies the change of heat field symmetry and heat field rotation through the spatial-temporal commutation of vertically aligned heaters. HFRM is actually a contact-free technique and can be applied for growing crystals in “closed” systems, e.g. with ampoule and hydrothermal methods [6].

The pioneering work on numerical modeling of convective heat-transfer in a rotating heat field was [7]. The previous paper [8] presented the numerical simulations of convective flows which exist in Ge and Si melts while growing
crystals in Czochralski configuration in a rotating heat field of second order axial symmetry \((\text{rotL}_2)\), i.e. when opposite heaters are successively switched in pairs.

This paper presents results of numerical modeling of convective heat-transfer in a rotating heat field of first order axial symmetry \((\text{rotL}_1)\), i.e. non-axial symmetric heat field [5]. We demonstrate flow patterns and temperature fields in a medium of crystallization with physical properties of Si and BSO melts, having several orders different Prandtl numbers.

2 Mathematical model

A sketch of the setup for crystal growth by Czochralski method and a calculated cell are shown in Fig. 1. A stationary mounted single crystal of radius \(R_c\) contacts the melt in a cylindrical crucible of radius \(R_0\) filled it up to level \(H_0\). The crucible is placed in a heating furnace with vertically aligned heaters. The average temperature inside the furnace is \(T_0\). The higher temperature of the sidewall of the crucible in a sector of \(H_0 R_0 \Delta \phi\) square is determined the temperature \(T_H\) of the opposite switched on heater \((T_H \geq T_0)\).

![Diagram](image)

Figure 1: a) The sketch of the setup of the contact - free heat field rotation method and a calculated cell; the filled circles are switched on heaters \((R_0 - \text{crucible radius}, H_0 - \text{liquid level in the crucible}, R_c - \text{crystal radius})\). b) The temperature distribution in a horizontal section - heat wave of \(\text{rotL}_1\) symmetry.

The mathematical modeling is based on the numerical solution of three-dimensional non-stationary Navier-Stokes equations in Boussinesq approximation and heat-transfer equations.

The assumption was made in considering a flat free surface of the liquid and favorable conditions for liquid adhesion on the crucible inner wall. Besides, at the initial moment no convection and/or other flows are suggested.
Non-compressible liquid movement equations are:

\[
U_t + \nabla(UV) = -\nabla P + \nabla^2 + kGr\theta, \quad U=(u,v,w)^T,
\]

\[
\nabla U = 0
\]

We add the heat transfer equation to system: \[\theta_t + \nabla(U\theta) = \frac{1}{Pr} \nabla^2 \theta \]

Where \(u, v, w\) are components of velocity vector in radial, azimuthal, and vertical directions, respectively; \(k\) is a unit vector along coordinate axis \(z\), \(P\) is the pressure, \(\theta\) is the temperature, \(Pr\) is the Prandtl number, \(Gr\) is the Grashoff number.

When going to dimensionless values we used the following characteristic parameters: \(R_0\) is the size, \(t_0 = R_0^2/\nu\) is the time, \(v_0 = v/R_0\) is the velocity, \(P_0 = \nu^2/\rho R_0^2\) is the pressure, \(\Delta T_0 = T_0 - T_s\) is temperature interval \((T_s\) is the temperature of phase transition, i.e. liquid solidification, \(\nu\) is the kinematic viscosity coefficient, \(\rho\) is the density). Dimensionless parameters are \(Pr = v/\alpha\), \(Gr = \beta g R_0^3 \Delta T_0/\nu^2\), \(Bi = \alpha R_0/\lambda\) (Biou number), where \(g\) is the free fall acceleration, \(\beta\) is the thermal volume expansion coefficient, \(\lambda\) is the thermal conductivity, \(\alpha\) is the thermal diffusivity, \(\nu\) is the convective heat exchange coefficient.

Dimensionless temperature is \(\theta = (T - T_s)/\Delta T_0\).

While determining the boundary conditions for the heat transfer eqn (3) we assume a complicated pattern of the heat exchange between the liquid in the crucible and environment, excluding the interfaces, where the temperature is constant. The maximum heating of the liquid tends to the areas close to the heaters. If we switch the heaters, the temperature in this part of the liquid would change continuously, i.e. during a certain time interval. New temperature values are determined by new heating conditions. To take this effect into consideration, the heat exchange between the liquid in the crucible and furnace environment is described by Newton’s law. Thus, the boundary conditions are stated as follows:

**Crucible bottom:** (0≤r≤1, \(z=0, 0≤\phi≤2\pi\)):

\[
u = 0, \quad \frac{\partial \theta}{\partial z} = Bi_1(\theta - 1),
\]

**Crucible sidewall:** \((r=1, 0≤z≤H \ (H = H_0/R_0), 0≤\phi≤2\pi)):

\[
\frac{\partial \theta}{\partial r} = \begin{cases} Bi_2(\theta_H - \theta), & \phi \in \Delta \phi_k, \\
Bi_2(1 - \theta), & \phi \notin \Delta \phi_k, \end{cases} \quad k = 0, 1, \ldots, K,
\]

Where \(\phi_k\) is the angle coordinate of the middle of the switched on heater, \(K\) is the number of heaters, and \(\Delta \phi\) is the width of a heated segment.

For a free surface \((z=H, 0≤\phi≤2\pi)):

\[
\frac{\partial u}{\partial z} = 0, \quad \frac{\partial v}{\partial z} = 0, \quad w = 0, \quad \frac{\partial \theta}{\partial z} = -Bi_3\theta, \quad \text{if } R_{cr} < r < 1, (R_{cr} = R_c/R_0),
\]
and \( u=0, \ v=0, \ w=0, \ \theta=0, \) if \( 0 \leq \rho \leq R_{cr}. \)

Initial conditions at \( t=0 \) are: \( u=0, \ v=0, \ w=0; \ \theta=1. \)

For the surface \( (0 \leq \rho \leq R_{cr}, \ z=H, \ 0 \leq \varphi \leq 2\pi) \ \theta=0. \)

### 3 Solution algorithm

Estimation of pressure is a part of the solution procedure. According to [9] and taking into account eqn (1), we derive an equation for \( P: \)

\[
- \nabla^2 P = \nabla[U_t + \nabla(UU)] - kGr\theta. 
\]  

(4)

Boundary conditions for \( P \) are defined from eqn (1). As at the bottom of the crucible \( z=0, \)

\[
\frac{\partial P}{\partial z} = -\frac{1}{r} \frac{\partial^2 P}{\partial \varphi^2} - \frac{1}{r} \frac{\partial^2 P}{\partial \varphi \partial \rho} + Gr\theta, \quad (r=1).
\]  

(5)

An analogous solution is used for the free surface \((z=H)\) and for the sidewall

\[
\frac{\partial P}{\partial z} = -\frac{1}{r} \frac{\partial^2 P}{\partial \varphi^2} - \frac{\partial^2 P}{\partial \rho \partial z}. 
\]  

(6)

Thus, systems eqns (4)-(6) is Neiman’s problem for Poisson’s equation, which should be solved for each time step.

For the solution of Navier-Stokes, Poisson and heat-transfer equations we used a finite-difference algorithm. We consider a regime of weak liquid convection implying the presence of laminar flows and the absence of a clear boundary layer. That is why the approximation of eqns (1), (3), and (4) was performed on an equidistant sparse grid. The difference scheme was described in details in [8].

Initially, the temperature field in the melt is calculated at each time step. The solution of the systems of algebraic equations - obtained by inexplicit approximation of eqn (3) - is performed by BSOR [10]. We used the velocity values in the convective members of eqn (3) from the previous time step.

The next step is the determination of pressure. After the approximation of eqn (4), to solve the obtained system we performed the expansion of the periodical grid function into a Fourier series at coordinate \( \varphi \) [11]. The solution of the system of algebraic equations for each coordinate of the angular variable is performed by incomplete factorization iteration method [10]. To determine the right part of eqn (4) we used the calculated velocity values from the previous time step and the determined temperature field. To avoid the accumulation of errors caused by the cessation of iterations by the convergence finite criterion we performed a correcting procedure like in [9], i.e. the approximation of eqn (4) implies that \( \nabla U^n = 0. \)

The substitution of the calculated temperature and pressure values into momentum equations would allow one to determine velocity field components. The solution of the algebraic system obtained by inexplicit approximation of eqn (1) is performed by BSOR. The velocity values in the convective members of
eqn (1) are taken from the pervious time step. Several iterations should be performed to correlate the distribution of pressure and temperature.

4 Results and discussion

4.1 Modeling of flow patterns in Si melt

The numerical experiments regarded heat field and flow patterns in a medium of crystallization with the properties of Si-melt produced by the additional heating of side walls with 12 vertically aligned heaters switched one by one (Fig. 1). Thus, we consider the conditions of a rotating heat field of first order axial symmetry - \( \text{rot}_{L_1} \). The 3-D diagram of temperature distribution is shown in Fig. 1b. Geometric parameters are: \( R_0=0.04 \) m, \( H_0=0.05 \) m, \( R_c=0.016 \) m, the width of each heated segment on the sidewall is \( \pi R_0/6 \) (\( \Delta \phi=\pi/6 \)). We used the following physical parameters for the Si melt \([12, 13]\): \( \lambda=67 \text{ Br/(m·K)} \), \( c_p=913 \text{ J/(kg·K)} \), \( \rho=2530 \text{ kg/m}^3 \), \( v=3.7\times10^{-7} \text{ m/sec} \), \( \beta=0.52\times10^{-5} \text{ 1/K} \), \( T_s=1688 \text{K} \). The temperature values are \( T_0=1693 \text{K} \) and \( T_{Hi}=1695+1698 \text{K} \). We used the following dimensionless parameters: \( Pr=0.013 \), \( Gr=1.2\times10^5 \), \( \theta=1+2 \), \( H=1.25 \), \( R_c=0.4 \), \( B_{i1}=3 \), \( B_{i2}=1 \), \( B_{i3}=0.1 \), and \( \Delta \phi=\pi/6 \) for the calculations, which were performed on a 20\( \times \)36\( \times \)25 sparse grid in radial, azimuthal and vertical directions, respectively. A time step of \( 10^{-4} \) was chosen according to the stability of numerical calculations and convergence of iterations.

Figs. 2 and 3 show the results of various schemes of non-symmetrical heating of the sidewalls. Figs. 2a and 2b illustrate the quasi-stationary distribution of the temperature field in the melt at \( \phi_1=180^\circ \) coordinate, when one heater is switched on and at \( \phi_1=1.5 \). It can be seen that the maximum heating tends to the area close to the heater, therefore, has no influence on the temperature distribution near the crystal (Fig. 2b). According to Fig. 2a the deviation of the minimal temperature from the central axis in the bottom part of the crucible is about 0.15.

Figs. 2c-2f show the flow pattern for this case. The circulation of the liquid is caused by the floating up of heat masses along the hot segments of the crucible vertical wall, their cooling near the surface and moving down along the central axis of the crucible. The strongest flow occurs near the central axis of the crucible, below the crystal, and near the heated segments of the side wall. The arrows in Fig. 2c show the direction of the flow and their length – the strength of the flow. Due to the non-symmetrical heating a part of the melt overcomes the descending flow along the central axis of the crucible and appears in the opposite side of the crucible. This is confirmed by the trajectories of the migration of marker-particles. The maximal temperature gradient in the azimuthal direction exists in the areas close to the switched on heater, resulting in the appearance of the flows radiating outward from the heater. Fig. 2d shows the flow pattern in the upper part of the melt (the circle in the central part of the figure shows the crystal outlines). A stable four-segment flow pattern, existing in the crucible, is symmetric about the plane going through the central axis of the crucible and the middle of the heater. It can be seen that the outline of the stagnant melt area
close to the crystal surface is significantly deformed by the flow moving from the switched on heater.

Figure 2: Distribution of temperature (a, b), velocity pattern (c, d) and marker motion trajectory (e, f) in the Si melt at $\theta_H=1.5$; a, c - in the $Oxz$ $\varphi=0^\circ$, $0 \leq r \leq 1$ and $\varphi=180^\circ$ $-1 \leq r \leq 0$ halfplanes; b, d in the $O\varphi$ plane at $z=0.975H$

The numerical simulations showed that if another heater is switching on, new stable circulation zones appear in the melt in a certain time interval. During this time period the area of the sidewall opposite to the formerly switched on heater is cooled (this process is enhanced by the convection), and the area close to the newly switched on heater is heated. The calculated trajectories of the markers and the results reported in paper [8] show that the distance between heaters and the time interval of their switching give a possibility for azimuthal melt flow along the zones of stable circulation.

This hypothesis is supported by Figs. 2e and 2f illustrating trajectories of particles under a counter-clockwise switching of heaters in $\Delta=0.05$ time intervals. The figures show the spatial trajectory of the particle and its projection onto the $O\varphi$ plane. The obtained results indicate that there is a stable azimuthal migration of a part of the melt following the direction of the switching. This migration appears in response to the synchronization of the switching of the heaters and the periods of the liquid circulation between the sidewalls and the central axis of the crucible. The average time of the revolution of marker-particles around the central axis of the crucible is 0.6 in the case under consideration, and it coincides with the $12\Delta$ period of heat field rotation. The calculations showed that flow patterns in the melt depend on the value of azimuthal temperature gradient, which is a result of uneven heating of the sidewalls. For example, at the $\theta_T=1.25$ temperature of the heater we failed to obtain a result even qualitatively similar to that in Figs. 2e and 2f; the trajectories...
of evenly distributed markers were within the 0rz plane. Analysis of the influence of $\theta_H$ on convective processes in the melt showed that at $\Delta=0.05$ a stable azimuthal flow, like appears at $\theta_H$ ranging from 1.4 to 1.8 (Figs. 2e and 2f).

Figure 3: Distribution of temperature (a, b), velocity pattern (c, d) and marker motion trajectory (e, f) in the Si melt at $\theta_H=2$; a, c - in the Orz $\varphi=0^\circ$, $0.5r\leq1$ and $\varphi=180^\circ$ -1$\leq$r$\leq0$ halfplanes; b, d in the Or$\varphi$ plane at $z=0.975H$.

Fig. 3 illustrates the results obtained at $\theta_H=2$. Figs. 3a and 3b show a quasi-stationary temperature distribution in the melt at $\varphi_r=180^\circ$ coordinate of the switched on heater. Qualitatively, the picture is the same as in Figs. 2a and 2b: maximum heating area is close to the heater; no influence of the local overheating on the temperature distribution near the crystal; in the bottom area the deviation of the minimum temperature from the central axis of the crucible is 0.2.

The flow pattern in the melt for this case is shown in Figs. 3c to 3f. As before, the main circulation is caused by the floating up of heat masses along the hot segments of the sidewall, they're cooling near the surface and moving down along the central axis of the crucible (Fig. 3c). Fig. 3d illustrates the character of the flow in the near-surface layer of the melt: the absence of the full symmetry is explained by the incompleteness of the process. Here, it is necessary to note that a significant reduction of the stagnant area near the crystal is caused by the increased intensity of the flow moving from the switched on heater.

Figs. 3e and 3f illustrate the migration of the market in the melt at the same regime of the switching of heaters, as in the above case. The figures show spatial trajectories of particles and their projections onto the $\varphi$ plane (symbols $s$ and $f$ are initial and final position of markers, respectively). According to the obtained results we can conclude that a counter-clockwise azimuthal circulation exists in the melt, however, it has different, sporadic character. The flows appearing due
to the strong heating of a segment of the side wall dominate (Fig. 3d), and there is no marker migration in this area within the $Orz$ plane. Such a circulation is possible only in that part of the melt, which is located at the opposite side of the crucible in respect to the switched on heater (Fig. 3c). This area remains stable before the next switching of the heaters. At $\theta_H=2$ we failed to obtain flow patterns similar to those in Figs. 2e and 2f at any value of parameter $\Delta$.

Figure 4: Distribution of temperature (a, b), velocity pattern (c), values of the component (d) and marker motion trajectory (e, f) in the BSO melt; a, c - in the $Orz$ $\varphi=45^\circ$, $0\leq r\leq 1$ and $\varphi=225^\circ$ - $1\leq r\leq 0$ halfplanes; b, d in the $Orz$ plane at $z=0.975H$.

4.2 Modeling of flow patterns in BSO melt

The possibilities of HFRM can be illustrated by the numerical experiments of flow patterns in the BSO melt-solution. Geometric parameters are: $R_0=0.025$ m, $H_0=0.03$ m, $R_c=0.01$ m, and the width of each heated segment on the sidewall is $\pi R_0/6$ ($\Delta \varphi=\pi/6$). The physical parameters for the $Bi_{12}SiO_{20}$ (BSO) melt solution are as follows [14]: $\lambda=0.345$ Br/(m·K), $a=1.15*10^{-7}$ m$^2$/sec, $\rho=7630$ kg/m$^3$, $v=3*10^{-6}$ m$^3$/sec, $\beta=7*10^{-5}$ 1/K, $T_0=1163$K. The temperature values are $T_0=1173$K and $T_H=2003$K. We used the following dimensionless parameters: $Pr=26$, $Gr=1.2*10^{10}$, $\theta_H=3$, $H=1.2$, $R_c=1.4$, $Bi_1=5$, $Bi_2=0.5$ and $\Delta \varphi=\pi/6$ for the calculations on the same sparse grid as considered above.

Figs. 4a and 4b show the quasi-stationary distribution of the temperature field in the melt produced at $\varphi=180^\circ$. It can be seen that the maximum heating tends to the area of the melt-solution free surface close to the heater. The thickness of this layer along the axis $z$ is small, and the overheating does not influence the temperature field near the crystal.

Figs. 4c and 4d show the flow pattern and the values of the component $v$ of the velocity vector, respectively. The calculations show that an azimuthal flow exists
near the crystal surface, and its intensity decreases as the depth increases. The azimuthal flows appearing in the near-surface layer of the melt-solution form a stable two-segment flow pattern. As far as the heating is asymmetric a part of the melt overcomes the descending flowing beneath the crystal and appears at the opposite half of the crucible. This is confirmed by the calculated trajectories of marker-particles.

Numerical simulations showed that new zones of stable circulation form in the melt in a certain time interval after another heater is switched on. Obviously, for a medium of crystallization with BSO properties the use of such parameters as the distance between the heaters and the time interval of their switching gives a possibility to perform re-distribution of the melt in the required direction along the zones of stable circulation. But the physical parameters of the BSO melt-solution (Pr > 1) are significantly different from those of Si melt (Pr << 1). So, initially we determined that the width of the heated segment on the sidewall must be not less than $\pi R_d/6$ and the parameter $\theta_H$ of the heater – not less than 3, i.e. the process requires a significant heating. We failed to obtain a result similar to that shown in Figs. 4e and 4f at less $\theta_H$ and width, because the azimuthal temperature gradient was very low.

As for the relationship between the time step $\Delta$ and convective processes in the melt-solution, a stable azimuthal flow pattern appears at $\Delta \geq 2$. Figs. 4e and 4f show a spatial trajectory of particles and its projections onto the $O\rho\phi$ plane, respectively, when the heaters are switched counter-clockwise at $\Delta = 2$ time intervals. The obtained results indicate that there is a stable azimuthal migration of the melt following the direction of the switching.

If the markers occur in the bottom area, they would be transported by the flow on a straight line between the center of the crucible and its sidewall. Then, while floating up, they start migrating toward the center. The attained values of the azimuthal component of velocity vector depend greatly on the distance between the marker and the liquid surface. Thus, if a particle appears in the area far from the sidewall and does not reach the upper layers of the melt-solution, it would move within the $O\rho\phi$ plane (the thickened parts of the trajectory in Figs. 4e and 4f). If a certain volume of the melt floats up along the sidewall, it would start migrating toward the center of the crucible just near the surface. The azimuthal deviation here is maximal. The time of the revolution of marker-particles around the central axis of the crucible is 24 at $\Delta = 12$ period of heat field rotation.

5 Conclusion

Using numerical simulations the authors studied the flows in the liquid at the non-symmetric cycle heating of the sidewalls of a cylindrical crucible by vertically aligned heaters. It was shown that the use of such heating parameters as the temperature of switched on heaters, the time interval of the switching, and the width of the hot segment of the sidewall of the crucible allows a contact-free control over thermo-gravitational convection in a medium of crystallization. This is true for the liquids with both low and high Prandtl numbers – Si melt and BSO
melts, respectively. Thus, the rotation of a heat field influences heat-mass transfer processes in melts and favors the destruction of a diffusive layer near crystal surface and even distribution of impurities in a grown crystal. The abandonment of the mechanical rotation of a crystal gives a possibility for the partial or full sealing of a growth crucible, increases the sterility of the processes and keeps the setup from possible vibrations.

References