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Numerical study of three-dimensional instabilities in a hydrodynamic model of Czochralski growth

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Abstract

A computational approach to the study of three-dimensional instabilities of flows in a Czochralski crucible is proposed. The flow is driven by buoyancy, thermocapillarity and rotation of the crystal and the crucible. The thermal boundary conditions account for the prescribed temperatures or heat flux, as well as convective and radiative heating or cooling of the boundaries. The numerical approach is based on finite volume discretization and consists of a Newton-type solver for the calculation of the steady flow states and an Arnoldi solver for the solution of the eigenvalue problems associated with the linear stability of the flow. Preliminary test calculations and examples of stability studies for the Czochralski melt flow are reported.

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1. Introduction

It is well known that the axial symmetry of the melt flow in a Czochralski crucible can be broken by an instability, which leads to three-dimensional non-stationary flow patterns, which, in their turn, impair the quality of a growing crystal. Prediction of the instability and understanding its physical mechanisms are necessary to find the means of flow control, which are capable of stabilizing the flow. However, this problem remains a challenge for computational modeling. Most numerical studies approach this problem by heavily CPU-time consuming three-dimensional unsteady computations (e.g., Refs. [1,2]), which hardly can provide the necessary answers when parametric analysis is needed. Another possibility is the three-dimensional stability analysis of a basic axisymmetric steady flow, which can be easily obtained as

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a numerical solution of the corresponding nonlinear axisymmetric problem. This leads to a generalized complex eigenvalue problem, which is the bottleneck for all the numerical analyses of such kind. Overcoming this difficulty allows one to perform an effective parametric study of possible flow instabilities and provide the necessary practical answers. In the case of crystal growth, for example, stabilizing the primary instability would mean stabilizing the process as a whole, which is extremely desirable for various crystal growth technologies.

In the present paper, we describe preliminary results on three-dimensional stability of axisymmetric flows in the Czochralski model and simplified flow models related to crystal growth processes. In the previously considered problems, where the onset of three-dimensional instability was studied, the flows were driven by convection or rotation (see Refs. [3–7] and references therein). These problems were solved mainly by the global Galerkin method with specially constructed nonorthogonal basis functions satisfying all the boundary conditions and the continuity equation [5]. This global Galerkin approach is well suited for flows in simple rectangular or cylindrical geometries, though it cannot be applied to much more complicated configurations of the crystal growth processes.

To approach practically important problems it is necessary to develop a stability solver for numerical methods based on a discretization of the flow region. Such a solver, developed for the finite volume discretization scheme, is described here. Similar stability solvers were described in several publications published during the last decade [5,8–10], but none of these solvers seem to be capable of handling the complicated crystal growth-related problems involving complicated geometries and nonlinear boundary conditions. The main blocks of the solver described below can be easily implemented for other discretization schemes.

In the following we briefly describe the problem and the methods used, report several comparisons between results obtained by the Galerkin and finite volume solvers, and show also examples of stability studies for the Czochralski crystal growth model for the parameters taken from Refs. [1,2].

2. Description of the problem and numerical method

The model of the Czochralski melt flow is sketched in Fig. 1. The flow region is a cylindrical crucible, which can be heated or cooled arbitrarily at its bottom and sidewall. The bottom and the sidewall are no-slip. The crystal and the crucible can rotate around their common axis. The central part of the upper boundary simulates the growing crystal. It is no-slip and can rotate independently from the crucible. Another part of the upper boundary is the melt surface, on which the thermocapillary force can act. The melt surface is assumed to be flat. Therefore the flow is driven by (i) buoyancy convection, (ii) thermocapillary convection, and (iii) rotation. Clearly, this is a simplified model, which does not account for many important phenomena, however allows us to study the main features of the flow instability.

The melt flow is described by the momentum, continuity and energy equations in the Boussinesq approximation. Assuming that the problem is completely axisymmetric and that the basic axisymmetric flow $\mathbf{U}(r,z), P(r,z)$ and $T(r,z)$ can be calculated, we consider the three-dimensional infinitesimally small perturbations of the basic state. The perturbations are defined as $\{\mathbf{u}(r,z), p(r,z), \tau(r,z)\}\exp[\mathrm{i}(k\theta+\lambda t)]$. The linear stability analysis leads to an eigenproblem for the time amplification rate λ. It is well known that the resulting eigenproblem is defined in the (r,z) plane and contains the azimuthal wave number k as an additional governing parameter [5].

Assuming that the flow region is mapped onto a certain grid and that the discretization at a grid node (r_i,z_j) is known, the generalized stability problem can be expressed in the following form:

$$\begin{aligned}\lambda\mathbf{u}_{ij} = &-[(\mathbf{u}\cdot\nabla)\mathbf{U}]_{ij}-[(\mathbf{U}\cdot\nabla)\mathbf{u}]_{ij}-[\nabla p]_{ij}\\ &+[\Delta\mathbf{u}]_{ij}+Gr\tau_{ij},\\ [\nabla\cdot\mathbf{u}]_{ij}=0,\quad \lambda\tau_{ij} = &-[(\mathbf{U}\cdot\nabla)\tau]_{ij}-[(\mathbf{u}\cdot\nabla)T]_{ij}\\ &+Pr^{-1}[\Delta\tau]_{ij}.\end{aligned}\tag{1}$$

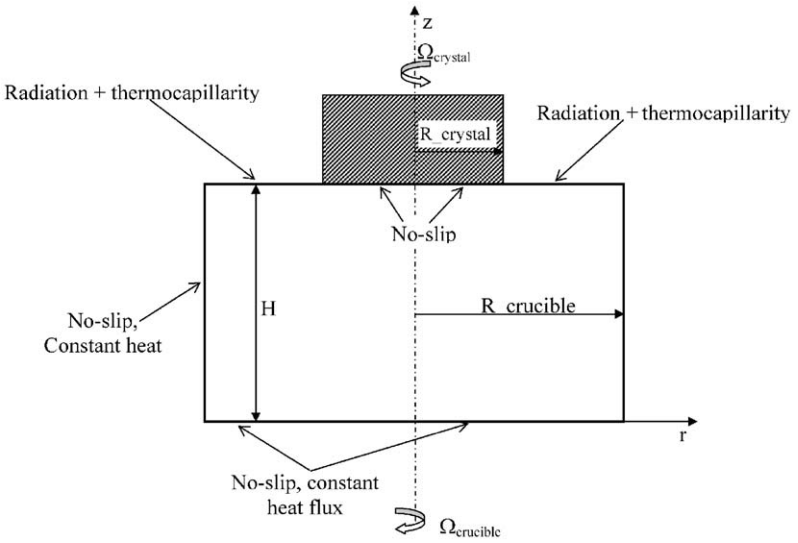

Fig. 1. Sketch of the problem.

Here $[\cdot]_{ij}$ denote the discretization at a node, Gr and Pr are the Grashof and the Prandtl numbers, respectively. Eq. (1) apply to all the inner nodes. Additional equations describing the boundary conditions must be supplied in all the boundary nodes. These equations can contain thermocapillary forces, rotation of the boundaries (i.e., of the crystal or the crucible), as well as the terms describing heating or cooling of a certain boundary.

The complete set of the linearized equation leads to the generalized eigenproblem

$$A\mathbf{x} = \lambda B\mathbf{x}, \tag{2}$$

where $\mathbf{x}$ is the vector of unknowns, and A and B are complex matrices. Due to the continuity equation and the boundary conditions the matrix B is singular, so that problem (2) cannot be transformed into a standard eigenvalue problem.

The whole computational process is separated into two main blocks. The first block yields the steady axisymmetric base state solution and the second one computes several leading eigenvalues of the linearized stability problem. In our codes the base state solution is calculated by the Newton iteration with parameter continuation where necessary. The Newton method is formulated in two versions: Jacobian-full and Jacobian-free. Each Newton iteration needs a solution of systems of linear equations, which is solved by the BICG(2)-stab algorithm. Our experience shows that the calculation of steady state, even for complicated cases, does not cause significant problems if a proper parameter-continuation is chosen. The iterative solver can be replaced or optimized for a certain problem, however a possible speedup seems to be negligible compared with the CPU time consumed by the eigenvalue solver.

The eigenproblem (2) is solved by the Arnoldi iteration in the shift-and inverse-mode

$$(A - \sigma B)^{-1} B\mathbf{x} = \mu\mathbf{x}, \quad \mu = 1/(\lambda - \sigma), \tag{3}$$

where σ is a complex shift. It should be noted that this approach succeeds when the shift σ, which can be complex, is chosen close to the leading eigenvalue λ. It is an easy task for benchmark problems, where the estimate of λ is known. However, it is an additional difficulty for each new problem where no information on the stability properties of the flow is available.

Each Arnoldi iteration requires the solution of the linear equations system $(A - \sigma B)\mathbf{x} = \mathbf{b}$. The usual approach is an iterative solution of these equations. The iterative solution usually requires too many iterations, because the right-hand side vector $\mathbf{b}$ changes completely from one iteration to

another, so that no good initial guess for the solution can be supplied. We have realized another approach, in addition to the iterative one, which builds the LU decomposition of the sparse matrix $(A-\sigma B)$. This consumes much more computer memory and a certain amount of CPU time for calculation of the LU decomposition. At the same time, the consequent Arnoldi iterations become fast, which allows us to calculate quite many leading eigenvalues. The number of the eigenvalues in different runs varies from 10 to 100. This approach can fail when the matrix is ill-conditioned. Note, that the iterations can diverge as well, so that two different linear solvers give us a possibility to attack more problems.

To validate the developed code we consider a series of test problems for the flows driven by buoyancy convection only, or thermocapillary convection only, or by rotation only. Tables 1–3 report the corresponding comparisons with the independent calculations of Refs. [4,6,11]. Details on the problem formulations can be found in the cited papers. The satisfactory results of the comparison allow us to continue the study for more complicated flows in the Czochralski configuration.

3. Results for Czochralski configuration

For the preliminary calculation we studied the flow of the melt of $LiCaAlF_6$ ($Pr = 1.4$) considered in Ref. [1]. In this case the crucible sidewall and the crystal surface are isothermal, the melt upper surface, the crucible bottom are thermally insulated. The crystal rotates with constant angular velocity and the crucible is motionless. As noted above, the flow is driven by buoyancy, thermocapillarity and rotation. Other details and parameters can be found in Ref. [1]. Since both

Table 1
Critical parameters for buoyancy convection in a cylinder with parabolic heating from the side

	Finite volume, 90×90 grid, present calculation		Galerkin method, 30×30 functions, result of Ref. [4]	
	Gr_{cr}	$\omega_{cr}/\sqrt{Gr_{cr}}$	Gr_{cr}	$\omega_{cr}/\sqrt{Gr_{cr}}$
$k=0$	512,964	0.4224	513,157	0.4600
$k=1$	325,625	0.2623	324,920	0.2614
$k=2$	96,254	0.1970	95,851	0.1977
$k=3$	312,483	0	318,059	0
$k=4$	312,612	0	311,111	0
$k=5$	810,000	0	809,618	0

$H/R = 2, Pr = 0.03$.

Table 2
Critical parameters for swirling flow in a cylinder with rotating lid

	Finite volume, 100×100 grid, present calculation		Galerkin, 30×30 functions, result of Ref. [6]	
	Re_{cr}	ω_{cr}	Re_{cr}	ω_{cr}
$k=0$	2564	0.2336	2584	0.23486
$k=1$	3308	0.06803	3308	0.06828
$k=2$	2993	−0.01615	2996	−0.01592
$k=3$	3919.5	−0.1112	3919	−0.1116
$k=4$	5890	−0.1988	5939	−0.2000
$k=5$	6767	−0.8353	6586	−0.8334

$H/R = 2$.

Table 3
Critical parameters for thermocapillary convection in a cylindrical liquid bridge

Pr	k_{cr}	Finite volume, 100×100 grid, present calculation		Result of Ref. [11]	
		$Mn_{cr} = Pr\,Ma_{cr}$	ω_{cr}	$Mn_{cr} = Pr\,Ma_{cr}$	ω_{cr}
0	2	1784	0	1793	0
0.01	2	1896	0	1901	0
0.02	2	2057	0	2062	0
0.05	2	3520	0	3522	0
0.06	3	13,270	171	13,251	179
0.07	2	18,319	54.8	18,302	54.3
0.08	2	17,384	66.2	17,356	65.7
0.1	2	16,130	74.9	16,094	74.2
0.16	2	14,530	75.2	14,504	75.0
0.2	3	13,290	396	13,275	393
0.3	3	10,531	312	10,545	317
0.7	3	7575	187.4	7570	187
1.0	2	2533	64.2	2551	65.0
2.0	2	1397	40.1	1407	40.1
4.0	2	997	28.3	1002	28.5

H/R = 1.

buoyant and thermocapillary forces are created by the temperature gradient, we considered the difference between the crystal and the sidewall temperatures ΔT as a critical parameter. Using the thermophysical properties of $LiCaAlF_6$ [1] the Grashof and Marangoni numbers can be recalculated as

$$Gr = g\beta R_{\text{crucible}}^3 \Delta T/v^2 = 6.16 \times 10^3 \Delta T$$

and

$$Ma = -\gamma R_{\text{crucible}} \Delta T/\rho v\kappa = 92.26\Delta T.$$

Fig. 2 shows how the critical temperature difference varies with the rotation of the crystal. The lines correspond to the marginal instability of the most critical modes corresponding to the azimuthal wave numbers $k = 0, 1, 2$ and 3. To determine the stable region the left and lower envelope of all the curves should be considered. It is seen that at low values of the crystal angular velocity $Re_{\text{crystal}} < 400$, $Re_{\text{crystal}} = \Omega_{\text{crystal}} R_{\text{crucible}}^2/v$, the melt flow remains stable at rather large temperature differences, $\Delta T > 100\,\text{K}$. When the crystal Reynolds number exceeds the value of 400 the critical temperature difference steeply drops below 20 K. Zeng et al. [1] reported that at $\Delta T = 50\,^\circ\text{C}$ the instability sets in beyond $\Omega_{\text{crystal}} = 15\,\text{rpm}$ with $k = 1$, which corresponds to $Re_{\text{crystal}} \approx 490$. According to the present results the instability sets in due to an axisymmetric perturbation at slightly lower angular velocity. Possibly, the three-dimensional pattern found in Ref. [1] corresponds to the secondary instability of the oscillatory axisymmetric motion.

For another example we considered the configuration of Ref. [2]. In this case the melt flow of $LiNbO_3$ ($Pr = 13.6, T_{\text{melting}} = 1526\,\text{K}$) is considered. The crucible sidewall is heated by a constant heat flux $q \approx 10^4\,\text{W}$ and the upper surface is cooled by the radiation to the enclosure, whose temperature is $0.8T_{\text{melting}}$. The present calculations, performed for the parameters of Ref. [2], show that the steady axisymmetric flow is unstable to three-dimensional perturbations with the azimuthal wave number varying from 0 to 13. Detailed results are not presented here due to lack of space. Thus, we expect that the pattern of the resulting three-dimensional flow will depend strongly on the initial conditions. The full unsteady three-dimensional calculations of Ref. [2] resulted in a three-dimensional pattern with fivefold azimuthal symmetry ($k = 5$). Our stability

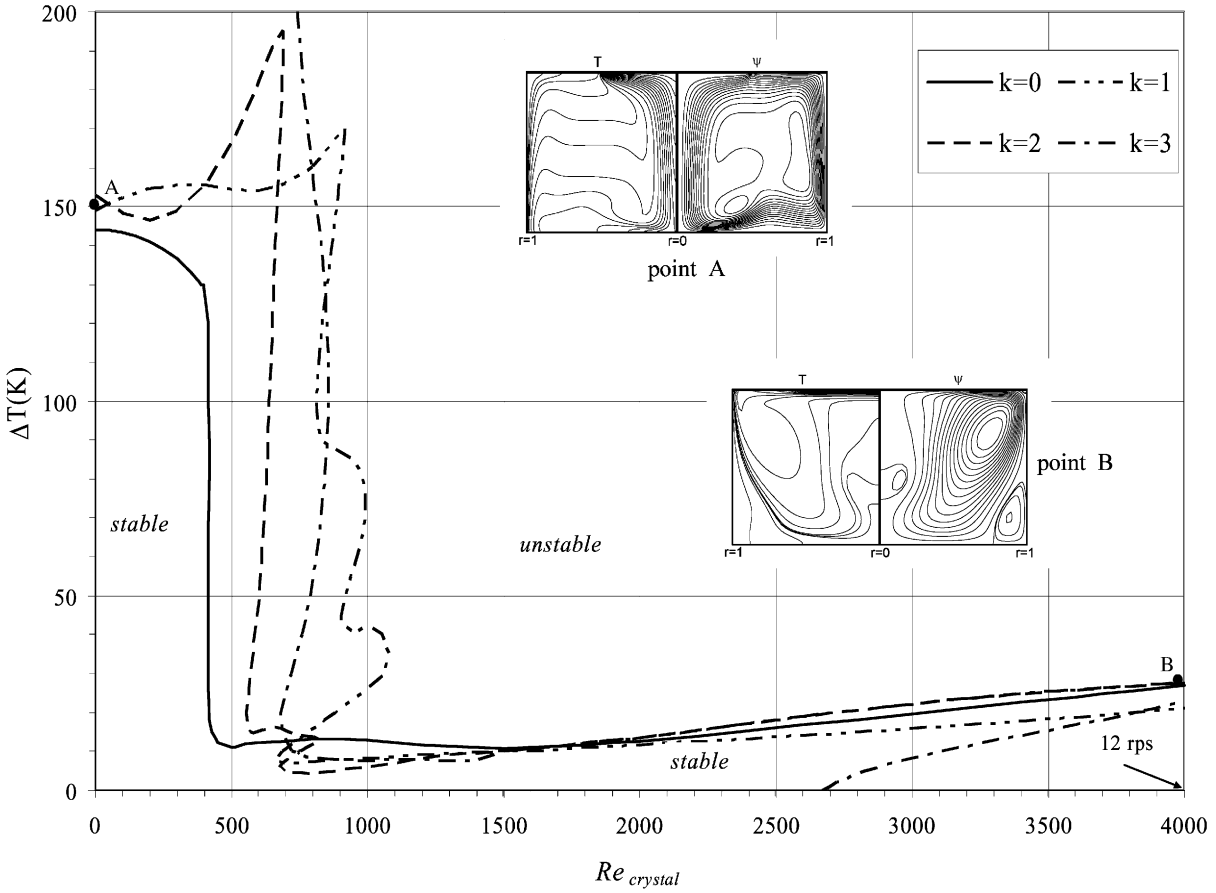

Fig. 2. Stability diagram of the melt flow in the Czochralski configuration considered in Ref. [1].

calculations show that this mode has the largest growth rate. However, we expect that patterns with other azimuthal symmetry could be found if other initial conditions will be specified.

Patterns of the fastest growing perturbations for the configuration of Ref. [2] show that the instability sets in at the free melt surface. The instability is caused by the thermocapillary force, which is large because of a steep temperature gradient there. The steep temperature gradient, in its turn, is the consequence of the heating from the side and cooling from the above. The heating cannot be decreased since then the melt temperature would decrease below the melting point. However, the temperature gradient and, consequently, the thermocapillary force can be decreased if the cooling of free surface will be weakened. This can be done, for example, by increase of the ambient temperature. Our calculations show that in the case of stationary crystal and crucible an increase of the ambient temperature from $0.8T_{\text{melting}}$ to $0.95T_{\text{melting}}$ would stabilize the flow drastically, so that the basic axisymmetric flow would remain stable even if the wall heat flux is tripled. With the increase of the angular velocity of the crystal the instability sets in abruptly similarly to the case illustrated in Fig. 2. Apparently, the comprehensive analysis of stability properties of this flow requires a detailed parametric stability analysis, which should result in a stability diagram showing, for example, the dependence of the critical wall heat flux on the crystal rotation for different ambient temperatures.

4. Concluding remarks

The present paper reports some preliminary results of the study of three-dimensional instabilities of an axisymmetric melt flow in a hydrodynamic model of Czochralski growth. It is shown that the numerical approach developed is capable of reproducing the previously published results obtained for simplified models, in which we studied the instabilities driven by the sole action of buoyant, thermocapillary and centrifugal/Coriolis forces. A rigorous validation of the developed approach for the Czochralski model still is needed. Unfortunately, the published three-dimensional calculations are performed on rather coarse grids and they cannot be used as reliable reference points.

Further development of the numerical approach described would include the heat transfer calculation in the growing crystal together with the calculation of the front of crystallization and the capillary meniscus. In case of semiconductor crystals the electromagnetic flow control should be also accounted for.

It should be emphasized that the hydrodynamic Czochralski model described is chosen because of its higher complexity compared to other methods of crystal growth from melts. The present numerical approach can be easily transformed to the study of other configurations, e.g., Bridgman or floating zone techniques.

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