Stability and bifurcation analysis for crystal growth processes

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Summary

The computational approach to the study of three-dimensional instabilities of flows related to bulk crystal growth from melts is described. The flow in a hydrodynamic model of the Czochralski crystal growth is taken as the target problem. Preliminary test calculations and an example of instability in the Czochralski system are reported.

Introduction

Most technological processes of crystal growth from bulk of the melt are carried out in axisymmetric configurations. However, instabilities of the melt flow usually lead to three-dimensional effects (e.g., spoke patterns), which strongly affect the quality of the growing crystals, and therefore are strongly undesirable. It is necessary, therefore, to predict the appearance of instability, to understand its physical mechanisms, and to find the flow control means capable of stabilizing the flow. The problem itself, when approached by computational fluid dynamics tools, is very complicated. As a rule, three-dimensional unsteady solvers are called for, which lead to heavily CPU-time consuming computations and can hardly provide the necessary answers when parametric analysis is needed. Another possibility is the three-dimensional stability analysis of an axisymmetric basic state flow, which is obtained as 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 a 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.

The present contribution describes our previous and novel results on three-dimensional stability of axisymmetric flows in the crystal growth related configurations. In the model, problems considered the flows that were driven by buoyancy convection, thermocapillary convection or rotation (see [1-5] and

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references therein). These problems were solved mainly by the global Galerkin method with specially constructed non-orthogonal basis functions satisfying all the boundary conditions and the continuity equation [3]. The global Galerkin approach is well suited for flows in simple rectangular or cylindrical geometries, though cannot be applied to much more complicated configurations of the crystal growth processes.

To approach the practically important problems, we started to develop a stability solver for numerical methods based on the discretization of the flow region. At the moment, most of our knowledge gained is for the finite volume discretization, but there are also some results obtained by the spectral element method. Similar stability solvers were described in several publications published during the last decade (not cited here due to the lack of space), but none of these solvers seem to be capable of handling the complicated crystal growth related problems.

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 [6].

Description of the problem and methods of solution

We consider the momentum, continuity and energy equations describing the non-isothermal motion of Newtonian incompressible fluid in the Boussinesq approximation. Assuming that the problem is completely axisymmetric and the basic axisymmetric flow $\boldsymbol{U}(r,z)$, $P(r,z)$ and $T(r,z)$ can be calculated, we consider the three-dimensional infinitesimally small perturbations of the basic state. The perturbations are defined as $\{\boldsymbol{u}(r,z),p(r,z),\tau(r,z)\}\exp[i(k\theta+\lambda t)]$. The linear stability analysis leads to the eigenproblem for the time increment λ. It is a well established fact that the resulting eigenproblem is defined in the (r,z) plane and contains the azimuthal wavenumber k as an additional governing parameter.

Assuming that the flow region is discretized by a certain grid and that the discretization in a node (r_i,z_j) is known, the stability problem has the following form

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

Here $[\cdot]_{ij}$ denote the discretization in a node, Gr and Pr are the Grashof and the Prandtl numbers respectively. Equations (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 thermophysical parameters describing heating or cooling of a certain boundary.

Obviously, the complete set of the linearized equation leads to the generalized eigenproblem

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

where **x** is the vector of unknowns, and A and B are the matrices. Due to the continuity equation and the boundary conditions the matrix B is singular, so that problem (2) cannot be transformed into a regular 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 the 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

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

where σ is a shift. It should be noted that this approach succeeds when the shift σ, which must be a 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 needs the solution of linear equations system $(A - \sigma B)\ \mathbf{x} = \mathbf{b}$. The usual approach is an iterative solution of these equations. The iterative solution usually needs too many iterations, because the right hand side vector **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, thus making the Arnoldi iterations very fast. This approach can fail however, when the matrix is ill-conditioned. Note, that the iteration can diverge as well, so that two different linear solvers give us a possibility to attack more problems.

The target problem: hydrodynamic model of Czochralski crystal growth

For the forthcoming stability studies we choose the model of the Czochralski growth [6], sketched in Fig.1. The melt flow takes place in a circular

crucible, which can be heated or cooled arbitrarily at its bottom and sidewall. The bottom and the sidewall are no-slip. The crucible can rotate around its 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. 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 allowing us to study the main features of flow instability. This model is implemented in our newly developed code, and the test calculations are being carried out.

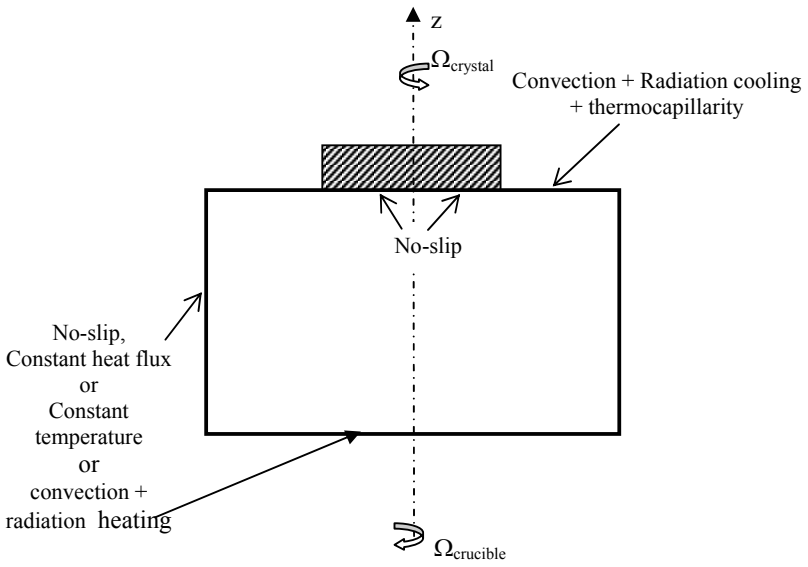

Fig.1. Sketch of the Czochralski hydrodynamic model

At the first stage of the test calculation we validate the flows driven by buoyancy convection only, or thermocapillary convection only, or only by rotation. For the buoyancy convection and rotation we have well validated results of [2,4] for the comparison. For the thermocapillary convection we compared with the results of [7,8] for the liquid bridges, which relates to the floating zone crystal growth technique. All the test calculations show that we can obtain correct critical values of the governing parameters, which are within 5% deviation with the previously published results, if the finite volume grid is fine enough. Usually we need more than 80 nodes of the stretched grid in one spatial direction to obtain the reasonable accuracy.

For the preliminary calculation we studied the configuration considered in [9]. In this case the crucible sidewall is heated by a constant heat flux and the upper surface is cooled by the radiation to the surrounding air. The governing parameters are: crucible aspect ratio $H/R_{crucible} = 1.957$, the relative crystal radius $R_{crystal}/R_{crucible} = 0.5$, the Prandtl number $Pr = 13.6$, the Grashof number $Gr = 2.62\times10^5$, the crystal Reynolds number $Re_{crystal} = 154.5$, the dimensionless sidewall heat flux $Q = 0.05$, and the upper surface radiation number $Rd = 0.34$. Further details are given in [9]. The axisymmetric basic state flow pattern for these parameters is shown in Fig.2. According to the results of [9] this flow is unstable and transforms into a 3D pattern with 5-fold azimuthal symmetry. This would imply the instability with the azimuthal wavenumber $k = 5$. Our calculations show that this mode is really the most critical, i.e. the real part of its time increment λ is the largest. The corresponding pattern of the temperature perturbation is illustrated in Fig.3. The stability analysis performed for different azimuthal modes showed that the flow is entirely unstable, also for modes with the azimuthal number k varying from 1 to 6. Further study showed that the configuration considered cannot be stabilized by a decrease of the value of the sidewall heat flux, since the "stable" value of Q would reduce the melt temperature below the melting point, which is obviously impractical. This flow possibly can be stabilized by other means, for example by the differential rotation of the crucible or by different heating of the boundaries. The described stability study is aimed to the search of such stabilizing conditions, which is the main goal of our forthcoming studies.

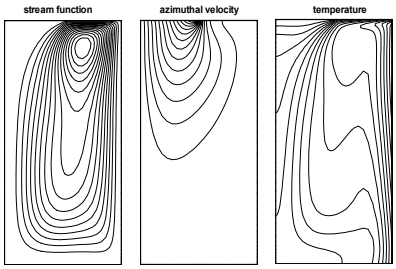

Fig.2. Pattern of the steady axisymmetric flow for the Czochralski growth problem considered in [9].

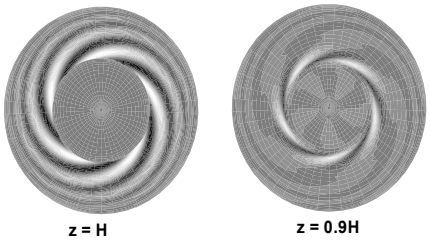

Fig.3. Isolines of the 3D perturbation of the temperature at the free surface and at the cross-section below it.

Concluding remarks

It should be emphasized that the hydrodynamic Czochralski model described is chosen because of its highest complexity among other methods of

the crystal growth from melts. Our numerical approach can be easily transformed to study other configurations, e.g., Bridgman or floating zone techniques. The external flow control means, such as electromagnetic devices, can be also accounted for rather easy.

For the future, work is planned also to implement the effects of phase change in the model, calculation of the shape of capillary interfaces and calculation of the temperature distribution in the growing crystal.

References

1. Gelfgat A.Yu., Bar-Yoseph P.Z. and Yarin A.L. (1999): Stability of Multiple Steady States of Convection in Laterally Heated Cavities. *Journal of Fluid Mechanics*, Vol. 388, pp.315-334.
2. Gelfgat A.Yu., Bar-Yoseph P.Z., and Solan A. (2000): Axisymmetry Breaking Instabilities of Natural Convection in a Vertical Bridgman Growth Configurations. *Journal of Crystal Growth*, Vol. 220, pp. 316-325.
3. Gelfgat A.Yu. (2001): Two- and Three-Dimensional Instabilities of Confined Flows: Numerical Study by a Global Galerkin Method. *Computational Fluid Dynamics Journal*, Vol. 9, pp. 437-448.
4. Gelfgat A.Yu., Bar-Yoseph P.Z., and Solan A. (2001): Three-Dimensional Instability of Axisymmetric Flow in a Rotating Lid - Cylinder Enclosure. *Journal of Fluid Mechanics,* Vol. 438, pp. 363-377.
5. Gelfgat A.Yu. and Bar-Yoseph P.Z. Multiple Solutions and Stability of Confined Convective and Swirling Flows – a Continuing Challenge. *Int. J. Numer. Meth. Heat and Fluid Flow*, to appear.
6. Hurle D.T.J. and Cockayne B. (1994): Czochralski Growth. In: Handbook of Crystal Growth, Elsevier Science, 1994, **Vol.** 2, pp. 99-211.
7. G. Chen, A. Lizee, and B. Roux, (1997): Bifurcation Analysis of the Thermocapillary Convection in Cylindrical Liquid Bridges, *Journal of Crystal Growth,* Vol. 180, pp.638-647.
8. M. Levenstam, G. Amberg, and C. Winkler (2001): Instabilities in Thermocapillary Convection in a Half-Zone at Intermediate Prandtl Numbers, *Physics of Fluids*, Vol. .13, pp.807-816.
9. C.J. Jing, N. Imaishi, T. Sato and Y Miyazawa (2000): Three-dimensional Numerical Simulation of Oxide Melt Flow in Czochralski Configuration", *Journal of Crystal Growth*, Vol. 216, pp. 372-388.