Parallel Finite Element Simulations of Czochralski Melt Flows

S. Adjerid, J.E. Flaherty, K. Jansen, M.S. Shephard

Scientific Computation Research Center
Rensselaer Polytechnic Institute
Troy, NY 12180-3590 USA

Summary

We use a finite element software based on the stabilized Galerkin Least-Squares method to solve the Navier Stokes equations with the Boussinesq approximation coupled to an energy equation through the temperature. We study melt flows associated with a Czochralski crystal growth process with the goal of understanding the transition from a steady laminar regime to an unsteady one as the Grashof number increases.

Introduction

Single crystals having specific electrical, mechanical, and optical properties are needed for high-performance electronic and opto-electronic applications involving, e.g., semiconductors and laser modulators. Mathematical modeling and numerical simulation have improved the ability to understand the bulk crystal-growth processes [1]. With powerful parallel computers, this can be advanced further to enhance and control these complex systems. The more limited goal of this investigation is an understanding of the transition from steady laminar to transient to turbulent flow of a molten liquid crystal with increasing Grashof number. The popular Czochralski (CZ) process [2] of bulk crystal growth features a rod holding an oriented seed crystal which is lowered through the top surface of the molten liquid contained in a crucible. With thermal control to maintain the upper surface of the fluid at the melt temperature, growth begins on the seed and when the crystal reaches a specified diameter, the rod is slowly withdrawn to continue growth.

The growth of the III-V compound indium phosphide (InP) crystals is much more difficult than, e.g., silicon. The CZ process is modified to include a viscous encapsulant (e.g., B_2O_3) on the free-surface of the melt to prevent phosphorous evaporation. The process is run at 40 atmospheres to further suppress the evaporation. The resulting technique is often called the modified liquid encapsulated Czochralski (LEC) process.

During processing, the flow undergoes Raleigh-Benard (buoyancy driven) convection and classical Benard (surface-tension driven) convection. Such flows may be characterized by the Grashof number, which is a ratio of the buoyancy to the viscous forces. As the Grashof number increases, axially-symmetric solutions cease to be stable and the flow bifurcates from steady states to unsteady states and eventually to turbulence [3].

Computational methods used to simulate crystal growth processes include finite difference [4], finite volume [5], finite element [6, 7], and spectral [8] methods. Herein, we use a Galerkin Least-Squares finite element method to solve the Navier-Stokes equations with the Boussinesq approximation.
with an energy equation coupled through temperature. We compute solutions on unstructured meshes of tetrahedral elements using a piecewise-linear finite element basis for all flow variables.

**Mathematical Model**

Consider the three-dimensional flow of an incompressible, viscous molten fluid in a cylindrical crucible of radius \( r_c \) and height \( H = r_c \). The planar bottom surface is joined to the vertical cylindrical surface by a quarter circular torus with radius 1mm. The top surface including the crystal-melt interface is assumed planar. The solid melt interface is assumed to be circular with radius \( r_s = r_c/2 \). The three-dimensional flow region is denoted as \( \Omega \). Its boundary \( \Gamma \) includes crucible walls \( \Gamma_c \), the melt-solid interface \( \Gamma_s \), and the encapsulant \( \Gamma_e \). The flow is governed by the Navier-Stokes equations and an energy equation [9]

\[
\rho_0 \frac{\partial \mathbf{u}}{\partial t} + \nabla \cdot (\mathbf{F}^{adv} - \mathbf{F}^{diff}) = \mathbf{S}, \quad \rho_0 c_p (T_t + \mathbf{u} \cdot \nabla T) - \nabla \cdot (\kappa \nabla T) = 0, \tag{1}
\]

where

\[
\mathbf{U} = [1, \mathbf{u}^T]^T, \quad \mathbf{F}^{adv}_i = \rho_0 u_i \mathbf{U} + p[0, \delta_{i1}, \delta_{i2}, \delta_{i3}]^T, \quad \mathbf{F}^{diff}_i = [0, \sigma_{1i}, \sigma_{2i}, \sigma_{3i}]^T, \tag{2}
\]

\[
\sigma_{ij} = \mu(u_{i,j} + u_{j,i}), \quad \mathbf{S} = [0, 0, 0, \rho_0 \beta g (T - T_0)]^T. \tag{3}
\]

Here \( \Gamma \mathbf{u} = [u_1, u_2, u_3]^T \) is the fluid velocity \( T \) is the temperature \( p \) is the pressure \( \sigma \) is the shear stress tensor \( \rho_0 \) is a reference density \( T_0 \) is a reference temperature (chosen as the melt temperature \( T_{mp} \) \( \beta \) is the thermal expansivity \( \mu \) is the viscosity \( c_p \) is the heat capacity \( \kappa \) is the heat conductivity \( g \) is the gravitational acceleration. A subscripted comma indicates partial differentiation with respect to time \( t \) or a spatial coordinate \( x_i \Gamma \hat{i} = 1, 2, 3 \).

The system of equations given by (1) is closed by imposing the initial conditions

\[
T = T_{mp}, \quad \mathbf{u} = 0, \quad t = 0, \tag{4}
\]

and the boundary conditions

\[
\mathbf{u} = 0, \quad \mathbf{x} \in \Gamma, \tag{5}
\]

\[
T = \begin{cases} 
T_c, & \mathbf{x} \in \Gamma_c \\
T_{mp}, & \mathbf{x} \in \Gamma_s 
\end{cases}, \tag{6}
\]

\[
q_c = -\kappa \partial_3 T_x = 0, \quad \mathbf{x} \in \Gamma_e. \tag{7}
\]
The no-slip condition at the melt-encapsulant interface is reasonable since the encapsulant is much more viscous than the crystal melt. With these conditions the temperature jumps from $T_{mp}$ to $T_c$ near the crystal-melt interface which leads to an abrupt change in the flow.

The system (1 - 7) is made dimensionless relative to the Grashof $Gr$ and Prandtl $Pr$ numbers

$$Gr = \frac{\rho_0 \beta gr_c^3 (T_c - T_{mp})}{\mu}, \quad Pr = \frac{\mu c_p}{\kappa}. \tag{8}$$

For large Grashof numbers the flow has viscous boundary layers of width $O(Gr^{-1/4})$ and thermal boundary layers of width $O(Gr^{-1/4}Pr^{-1/4})$ [9].

**The Finite Element Formulation and Solution**

When the standard Galerkin finite element method is applied to the incompressible Navier Stokes equations solutions exhibit spurious oscillations unless the mesh is sufficiently fine within layers. The stabilized Galerkin Least-Squares method [10] overcomes this difficulty while allowing the use equal-degree polynomial bases for all variables.[11].

While it is desirable to couple the energy equation to the fluid flow we currently determine the temperature in a staggered form. With this formulation we need only solve a scalar advection-diffusion equation for the temperature at each stagger step. Following Shakib [12] we introduce the solution vector $Y = [p, u_1, u_2, u_3]^T$ and define the quasilinear operator

$$\mathcal{L} \equiv A_0 \frac{\partial}{\partial t} + A_i \frac{\partial}{\partial x_i} - \frac{\partial}{\partial x_i} (K_{ij} \frac{\partial}{\partial x_j}) \tag{9}$$

where $A_0 = \rho_0 U \cdot Y$ is the variable-transformation metric $A_i = F_{iY}^{adv}$ is the $i$th Euler Jacobian matrix $K$ is the diffusivity matrix which is defined such that $K_{ij} Y_{,j} = F_{i}^{diff} [11]$ and repeated indices imply a summation.

Let $H^1(\Omega)$ denote the usual Sobolev space of functions having square-integrable first derivatives on $\Omega$; discretize $\Omega$ into a region $\Omega^e$ of $n_{el}$ finite elements; and define the trial space $V_h \subset H^1_0(\Omega^e)$ to consist of piecewise polynomials of degree $k \geq 1$ in each variable relative to $\Omega^e$ and to satisfy the essential boundary conditions (5 - 6). The test space $W_h \subset H^1_0(\Omega^e)$ must satisfy trivial essential boundary conditions. We construct a weak form of (1 - 7) to determine $Y \in V_h$ satisfying

$$0 = \int_{\Omega} \left(W \cdot U_t - W_{,i} \cdot F_{i,}^{adv} + W_{,i} \cdot F_{i,}^{diff} \right) d\Omega - \int_{\Gamma} W \cdot (-F^{adv} + F^{diff}) \cdot n d\Gamma$$

$$+ \sum_{e=1}^{n_{el}} \int_{\Omega^e} \mathcal{L}^T W \cdot \tau (L Y - S) d\Omega. \tag{10}$$

The first line of (10) contains the usual Galerkin statement of the problem and the second line contains the least-squares stabilization. We use the SUPG stabilization obtained by replacing $\mathcal{L}^T$ by $\mathcal{L}_{adv}^T$. The
stabilization matrix $\mathbf{\tau}$ is an important ingredient in these methods and is well documented by Shakib [12] and Franca and Frey [10].

Problem (10) leads to a nonlinear system of ordinary differential equations of the form

$$M\dot{\mathbf{Y}},_t = \mathbf{R}(\dot{\mathbf{Y}})$$

where $\dot{\cdot}$ indicates the discrete approximation of $\mathbf{Y}$. An implicit, second-order accurate family of time integration routines has been developed for this application. This new time integration software extends Chung and Hulbert’s [13] generalized alpha method. Time discretization results in a nonlinear algebraic problem that is solved as a predictor-corrector pair leading to successive linear algebraic problems. The solution of each linear system is notoriously difficult for incompressible flows, especially those at high Reynolds numbers and those associated with high aspect-ratio elements. We have found the commercial software ACUSOLVE to have a very robust algebraic procedure for the present formulation.

We solve (1-7) using an unstructured mesh of tetrahedral elements obtained by finite Octree techniques [14]. Meshes are graded near $\Gamma$ to resolve boundary layers [15]. Near the boundary $\Gamma\Gamma$ the mesh has $n$ layers of elements graded normal to $\Gamma$. Letting $r_i$ denote the thickness of layer $i$ $i = 0, 1, \ldots, n - 1$ where layer 0 is next to $\Gamma \Gamma r_i$ is given by

$$r_i = ar^i, \quad i = 0, 1, \ldots, n - 1,$$

with

$$r_0 = a \quad \text{and} \quad r_{n-1} = \sum_{k=0}^{n-1} ar^k = h.$$  

The mesh is isotropic elsewhere within $\Omega$. Graded meshes are required to resolve the flow and temperature in the boundary layers for Grashof numbers of half a million or higher.

**Numerical Computations and Discussion**

We solve the stabilized Galerkin problem (10) for LEC InP melt flows using the parameter values shown in Table 1. The mesh has six layers near the boundary with $n = 6\Gamma h = 0.05\Gamma$ and $a = 0.001$. There are a total of 634001 tetrahedral elements and 109994 vertices. We select a Grashof number of $6.610^6$ and solve (10) for 2000 time steps of duration 0.1 $\Gamma$ letting the Grashof number increase to $6.610^7$ linearly within a 10-second period $\Gamma$ and compute for 500 time steps of duration 0.1. We show velocity

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_c$</td>
<td>3.4 cm</td>
</tr>
<tr>
<td>$\rho_0$</td>
<td>5.05 g/cm$^3$</td>
</tr>
<tr>
<td>$\mu$</td>
<td>0.0081 g/cm - s</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>0.23 W/cm - K</td>
</tr>
<tr>
<td>$c_p$</td>
<td>0.42 J/g - K</td>
</tr>
<tr>
<td>$\beta$</td>
<td>4.44 x 10$^{-4}$</td>
</tr>
<tr>
<td>$T_{mp}$</td>
<td>1335 K</td>
</tr>
</tbody>
</table>

Table 1: Parameter values used for the LEC melt flow simulations.
vectors in horizontal and vertical-horizontal planes at \( t = 50 \) seconds in Figure 1. At this Grashof number the flow has transient fully three dimensional components. These calculations are preliminary. In subsequent analyses we hope to study the transition to turbulence including a turbulence model and the use of adaptive and parallel methods with dynamic load balancing [6].

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References