Control of Fluid Flow in the Vertical Bridgman Crystal Growth Process

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Abstract— **In this work we consider feedback control of flows in a vertical Bridgman crystal growth system, a process used to grow single crystals for a wide array of applications, ranging from lasers to high-speed microelectronics to infrared sensors. We study the feasibility of using proportional controllers to attenuate flow oscillations that occur in some operating regimes of the system. A spatial average of the flow speed is chosen as the single controlled output. This variable provides an effective scalar measure of the intensity and temporal variation of the hydrodynamics within the melt. The flows are controlled via rotation of the crucible containing the molten material. Simulations show that these simple feedback controllers are effective at stabilizing the flow.**

Keywords— **flow control, crystal growth**

I. Introduction

THE vertical Bridgman process is used to grow single crystals, for use as substrates in optoelectronic and sensing devices. To produce devices of suitable quality, it is necessary to grow low-defect single crystals of homogenous chemical composition. This task is made difficult by the complex coupling of heat, mass, and momentum transport inherent in the process. One processing issue that continues to challenge the crystal growth community is the occurrence of fine scale variations in composition, known as striations. These composition variations often cause undesirable variations in material and electronic properties. Crystal growth experiments by Kim, Witt, and Gatos [1] investigated the occurrence of striations in a thermally destabilized vertical Bridgman system. This configuration is very similar to the classical Rayleigh-Bénard problem, used to study the natural convection arising in a system that is heated from below and cooled from above. A Rayleigh number was used to measure the progress of crystal growth,

$$Ra_m = \frac{g\beta_T L_m^4 \zeta}{\nu \alpha_m} \tag{1}$$

where g is the magnitude of gravitational acceleration, β_T is the thermal expansivity, L_m is the melt height, ζ is the axial temperature gradient in the melt, ν is the kinematic viscosity, and α_m is the thermal diffusivity. It was found that above a critical Rayleigh number, the melt exhibited periodic, time-varying flows. It was then shown conclusively that these time-varying flows were directly related to the occurrence of striations.

In previous work [2] we used detailed crystal growth models to numerically simulate the experimental system of Kim, Witt, Gatos. Our models predict the occurrence of periodic, time-varying flows at Rayleigh numbers above a critical value. In the early stages of crystal growth, our numerical simulations predict the occurrence of large amplitude, two-frequency oscillations within the flow. In later stages of growth, the oscillatory behavior becomes more ordered with decreasing amplitude until finally the flows are steady with time. These simulations are in qualitative agreement with the experimental results of Kim, Witt, and Gatos. Demonstrating the appearance of periodic, time-varying flows in our model was an important first step in our study of crystal striations. The next step, the subject of this work, is the formulation and implementation of feedback control algorithms that will beneficially alter the fluid dynamical behavior within the system.

Flow control is a rapidly emerging field. A detailed discussion of the current applications, techniques and unresolved issues is beyond the scope of this paper. For a general review, see [3]. Here, we will only highlight a few of the more pertinent works. A good deal of work has been devoted to using feedback control to delay the onset of convection in Rayleigh-Bénard systems. Howle [4] used proportional control to stabilize the quiescent flow state for supercritical Rayleigh numbers in a 1-D Rayleigh-Bénard experiment. A network of heaters, installed at the bottom of the container, was used to stabilize the flows. Tang and Bau investigated the delay of convection in Rayleigh-Bénard systems in both experimental [5] and numerical [6] investigations. By using diodes to measure fluid temperature and micromachined heaters at the bottom of the test cell, the researchers were able to apply proportional control to stabilize 3-D flows for supercritical Rayleigh numbers. Shiomi and Amberg [7] applied proportional control to attenuate oscillations arising in a floating zone crystal growth system. Two sensor/heater pairs were used for measurement and actuation. They found that complete suppression of the oscillations could be achieved for weakly nonlinear flows but was not possible for stronger nonlinear flows.

In this work, we use crucible rotation in conjunction with proportional controllers to modify flows in the vertical Bridgman system. Open-loop crucible rotation has been successfully applied to experimental vertical Bridgman systems [8] and has been the subject of simulation-based studies (see [9] and references therein). Generally, the purpose of crucible rotation in crystal growth systems is to promote mixing of the melt. Here, we wish to use crucible rotation to suppress flow oscillations. When implemented in practice, this would be a significant step towards the removal of striations in crystal growth processes.

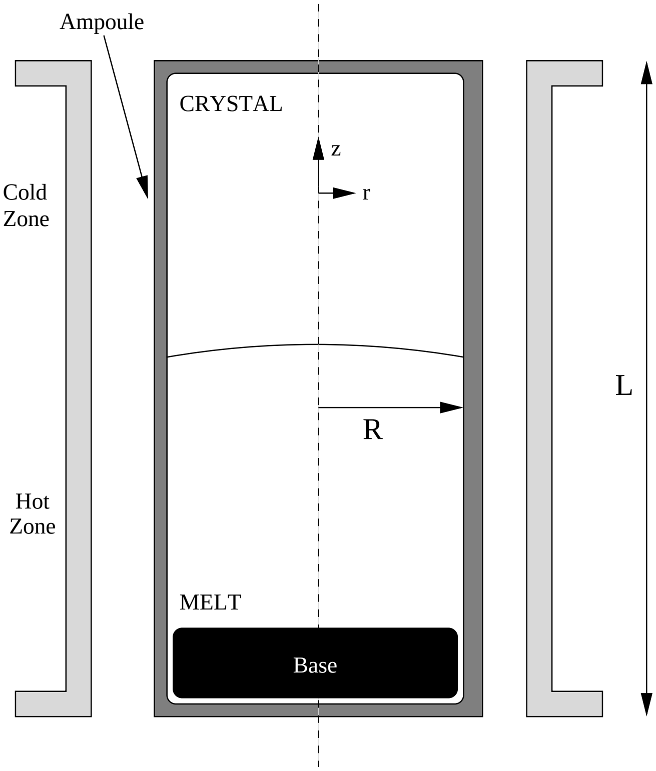

Fig. 1. Vertical Bridgman growth system

II. The Vertical Bridgman Process Model

A schematic of the vertical Bridgman process is shown in Figure 1. Polycrystalline material is loaded into a quartz crucible (sometimes called an ampoule), inside of a high temperature furnace. The material is completely melted, at which time the distribution of temperature in the furnace is adjusted to vary along its length, such that one zone is hotter than the material's melting point and the other section is cooler. Directional solidification is achieved by slowly translating the translating the crucible towards the cold zone of the furnace.

We use equations for conservation of mass, momentum, and energy, along with appropriate boundary and initial conditions, to model the vertical Bridgman system. Our present purposes only require that we study the effect of control on flow, so we do not include conservation of chemical species in the model, although doing so would be necessary for the direct prediction of striations. A key assumption used here is that the system is perfectly axisymmetric, in which case all field variables are independent of the azimuthal coordinate.

Under the these assumptions, the time-dependent energy transport equation is given by

$$Pr_m \frac{\partial T}{\partial t} + v \cdot \nabla T - \nabla^2 T \quad = \quad 0 \tag{2}$$

in the melt, and

$$Pr_j \frac{\partial T}{\partial t} - \nabla^2 T \quad = \quad 0 \tag{3}$$

where the index j designates the material, either the crystal, crucible, or graphite support. T is a dimensionless temperature scaled by the melting temperature, t is a dimensionless time scaled by R^2/ν, and ∇ is the dimensionless gradient operator, in which the spatial coordinates are scaled by the crucible radius R. The Prandtl number is defined as $Pr_i \equiv \nu/\alpha_i$, where ν is the kinematic viscosity, and the thermal diffusivity is defined as $\alpha_i \equiv k_i/\rho_i C_{p,i}$, where k_i is the thermal conductivity, ρ_i is the density, and $C_{p,i}$ is the heat capacity.

The velocity field is described by the Navier-Stokes (N-S) equations for incompressible flow, using the Boussinesq approximation to account for temperature-dependent density variation:

$$\nabla \cdot v = 0 \tag{4}$$

$$\frac{\partial v}{\partial t} + \frac{1}{Pr_m} v \cdot \nabla v - \nabla \cdot \mathsf{T} - Ra_T (T-1) e_z = 0 \tag{5}$$

Here $v(r,z)$ is the dimensionless velocity scaled by α_m/R, T is the stress tensor, and e_z is the unit vector in the axial direction. The stress tensor is split up into the dynamic pressure and the strain rate tensor,

$$\mathsf{T} = -P\mathsf{I} - \tau = -P\mathsf{I} + (\nabla v + \nabla v^T) \tag{6}$$

where P is the dynamic pressure scaled by $\rho_m \nu \alpha_m / R^2$, and I is the idemfactor.

A thermal flux condition is defined at the melt-crystal interface,

$$-(\kappa \nabla T|_m + \nabla T|_s) \cdot \mathbf{n_{sm}} = St Pr_s (\mathbf{n_{sm}} \cdot \dot{\mathbf{x}}) \tag{7}$$

where κ is the ratio of melt thermal conductivity to solid, $\kappa = k_m/k_s$, $\mathbf{n_{sm}}$ is the unit vector normal to the melt-crystal interface, pointed towards the melt, St is the Stefan number, $St = \Delta H_f / C_{p,s} T_{mp}$, and $\dot{\mathbf{x}}$ is the velocity of the melt-crystal interface.

The temperature at the melt-crystal interface is assumed to be equal to the material melting point,

$$T = 1 \tag{8}$$

where T is the dimensionless temperature. A linear furnace temperature profile used, which is a reasonable approximation of the furnace used in Kim, Witt, and Gatos' experiment:

$$T_f = T_c + \frac{(T_h - T_c)}{L}(z - z_o) \tag{9}$$

where T_f is the furnace temperature, T_c and T_h are the minimum and maximum furnace temperatures, respectively, and z_o is the furnace reference position. Convective and radiative heat transfer from the furnace to the crucible are represented by a simple flux condition applied at the domain boundary:

$$-\nabla T|_a \cdot n_{af} = Bi_i (T - T_f(z)) + Rd_i (T^4 - T_f^4(z)) \tag{10}$$

where the Biot number, $Bi_i = hR/k_i$, is a dimensionless heat transfer coefficient, the Radiation number, $Rd_i = \sigma\epsilon_i RT_{mp}^3/k_i$, relates radiative effects to conductive effects, ϵ is the crucible emissivity, σ is the Stefan-Boltzmann constant, and n_{af} is the unit vector normal to the crucible/furnace interface. No-slip boundary conditions are assumed at the crucible walls and melt-crystal interface,

$$v = \Omega r \mathbf{e}_\theta \tag{11}$$

where Ω is the crucible rotation rate and $\mathbf{e}_\theta$ is the unit vector in the azimuthal direction. Finally, at the centerline of the system, symmetry conditions are assumed,

$$\frac{\partial T}{\partial r}|_{r=0} = 0 \tag{12}$$

$$v \cdot e_r|_{r=0} = 0 \tag{13}$$

$$\frac{\partial (v \cdot e_z)}{\partial r}|_{r=0} = 0 \tag{14}$$

The governing equations that comprise the KWG model are nonlinear, coupled, partial differential equations. The Galerkin Finite Element Method (GFEM) was employed to spatially discretize equations for energy transport (Eq. 2), momentum transport (Eq. 5), and mass continuity (Eq. 4). This system of nonlinear DAEs was time integrated using a second-order implicit trapezoidal method, and the resulting system of nonlinear algebraic equations was solved using a modified Newton's method. A dimensionless time step of $\Delta t = 0.01$ was shown to provide satisfactory numerical convergence as well as sufficient resolution of the time-varying phenomena within the system. A single factorization of the problem with a discretization of 25592 degrees of freedom required approximately 4.8 seconds on a mid-priced PC with a processor speed of 2.0 GHz . For the interested reader, additional details of the model and numerical scheme can be found in [2], [10].

III. The Control Problem

Our general research objective is the application of feedback controllers to control flows within our detailed model of the vertical Bridgman process. One particularly important application is the stabilization of flow oscillations to prevent the occurrence of crystal striations. In previous work [2], our simulations of the Kim, Witt, and Gatos experiment indicated the existence of periodic, time-varying flows at sufficiently high Rayleigh numbers. The onset of periodic behavior was shown to correspond to a Hopf supercritical bifurcation occurring at a critical Rayleigh number, $Ra_{cr} = 3.57 \times 10^5$. The spatial average of speed, referred to from this point on simply as speed, provides a global measure of the flow intensity, and can be used to characterize the time-dependent behavior of the flow:

$$s = \sqrt{\frac{1}{V}\int_V (v_r^2 + v_z^2)\, dV}, \tag{15}$$

where v_r and v_z are the respective radial and axial components of the velocity field and V is the volume of the melt region. Figure 2 shows the spatial average of the speed as a function of time for $Ra = 3.6 \times 10^5$. The figure shows that the flows exhibit periodic, time-varying behavior, with oscillations of single period and small amplitude, at this supercritical value of Rayleigh number.

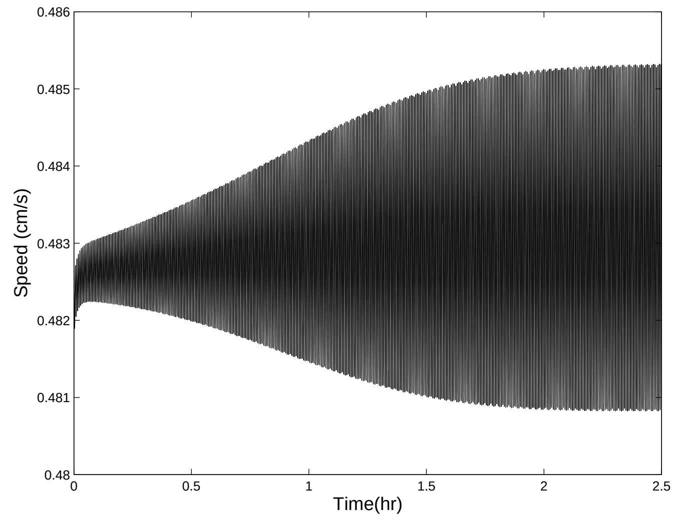

Fig. 2. Open-Loop Response for $Ra = 3.6 \times 10^5$

There are many actuation techniques that could perceptibly affect the fluid dynamics, and be feasibly implemented, in this crystal growth system. Some possibilities include real-time adjustment of the thermal environment, either by changing the furnace thermal profile or providing a localized heater, application of vibration, or use of a magnetic field. This work addresses the use of crucible rotation (rotation rate, Ω) as the manipulated input. In industrial practice, both steady rotation and time-dependent rotation have been used. Time-dependent rotation is most often implemented in a manner described as the Accelerated Crucible Rotation Technique (ACRT) [11]. ACRT rotates the crucible according to a pre-determined schedule: the purpose is to use alternating acceleration and deceleration to drive secondary flows that improve mixing in the melt.

Open-loop simulations were conducted to better understand the effect of crucible rotation on flows within this system. Steady-state calculations were conducted for $Ra = 3.6 \times 10^5$, with steady crucible rotation rate, Ω, varied from zero to 4.4 rpm. The speed computed from these results is shown in Figure 3. As seen in the figure, with all other parameters held constant, there is a one-to-one correspondence between rotation rate and flow speed. An increase in rotation rate suppresses the radial and axial components of the velocity field, and consequently the flow speed, with a limiting behavior of solid-body rotation at very high rotation rate (although this limit cannot be achieved in practice, due to onset of three-dimensional flow instabilities).

These observations motivate the question as to whether open-loop steady rotation can be successfully used to attenuate flow oscillations. Our simulations show that the

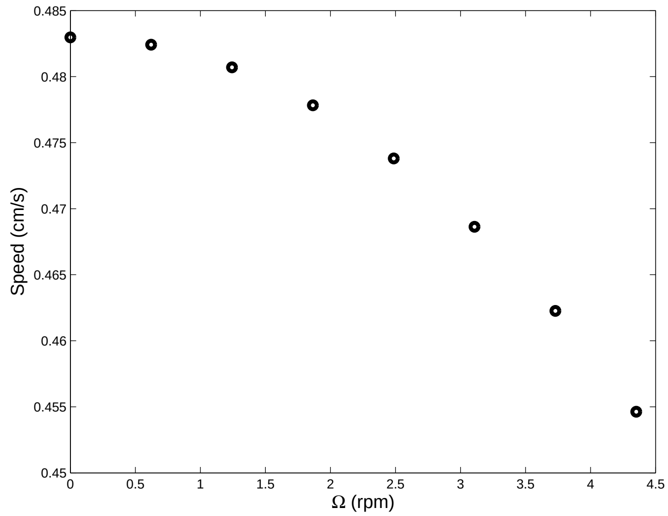

Fig. 3. Flow Speed vs. Rotation Rate: Steady State Results

opposite is true: steady rotation exacerbates the flow oscillations. Figure 4 presents results that demonstrate this conclusion, obtained using transient simulations for $Ra = 3.6 \times 10^5$. Steady states are denoted as circles, flanked by triangles that represent the maximum and minimum amplitudes of the oscillations. It is clear that as the steady rotation rate is increased, the amplitude of oscillations also increases. Thus, while steady crucible rotation affects the flow, it does not attenuate the periodic flow oscillations that occur at high Rayleigh numbers.

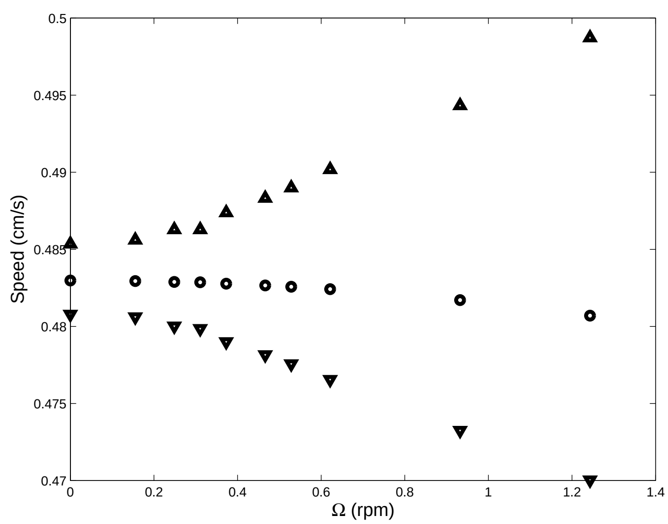

Fig. 4. Flow Speed vs. Rotation Rate: Transient Results

We now address the feasibility of using feedback control to suppress flow oscillations. The controlled output is chosen to be the spatial average of the kinetic energy, E, a scalar quantity that captures the essential dynamics of the flows within the process. The kinetic energy is simply the speed (defined in Eq. 15) squared. All states are assumed to be measurable and are available from the vertical Bridgman process model. We apply a proportional controller,

$$\Omega(t) = K_c(E_{sp} - E(t)) \tag{16}$$

where K_c is the controller gain and E_{sp} is the kinetic energy setpoint.

IV. Results and Discussion

This section presents results from our closed-loop simulations of the vertical Bridgman process. We apply proportional control to our model of the Kim, Witt, and Gatos experiment for $Ra = 3.6 \times 10^5$. The initial condition is a solution obtained from the open-loop transient simulations once the flow has fully evolved to its time-periodic state. In all of the simulations, the setpoint was set to $0.2284\ \mathrm{m}^2/\mathrm{s}^2$, equivalent to a speed $= 0.478$ m/s. Since the application of crucible rotation reduces the kinetic energy of the flow, the setpoint must be chosen at a kinetic energy value less than the steady state kinetic energy corresponding to no rotation $\Omega = 0$. The data in Figure 3 indicate that this setpoint value is physically reasonable.

Figure 5 shows the speed as a function of time, for $K_c = 0.75$. It is clear that proportional control is successful in attenuating the flow oscillations. As seen in the figure, the final value of the speed is within one percent of the setpoint. Figure 6 provides a closer view of the time-variation of speed. The flow oscillations have a period of approximately 20 seconds, which is consistent with both open-loop experiments [1] and open-loop simulations [2].

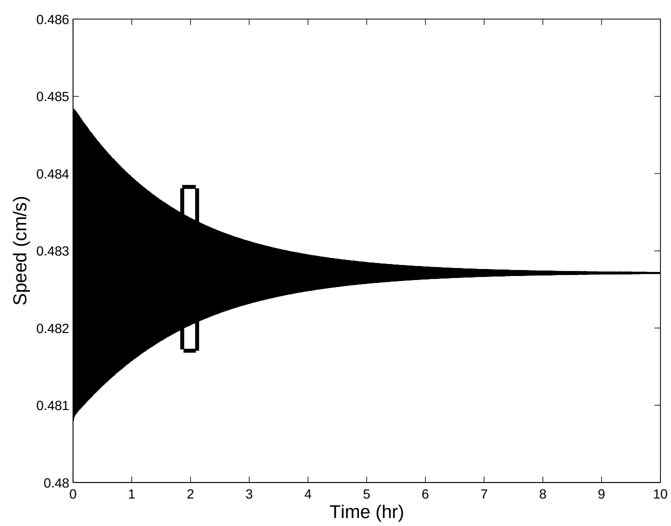

Fig. 5. Flow Stabilization: Speed vs. Time, $K_c = 0.75$

Corresponding to the same simulation, the crucible rotation rate is given as a function of time in Figure 7. The crucible only rotates in one direction, but speeds up and slows down in response to the inherent oscillations in the flow. Since the crucible rotation rate is less than 1 rpm, this control scheme is feasible from a practical engineering point of view. A close-up of the rotation rate, corresponding to the same time window as Figure 6, is given in Figure 8. As expected, the oscillations in rotation rate follow the flow oscillations closely.

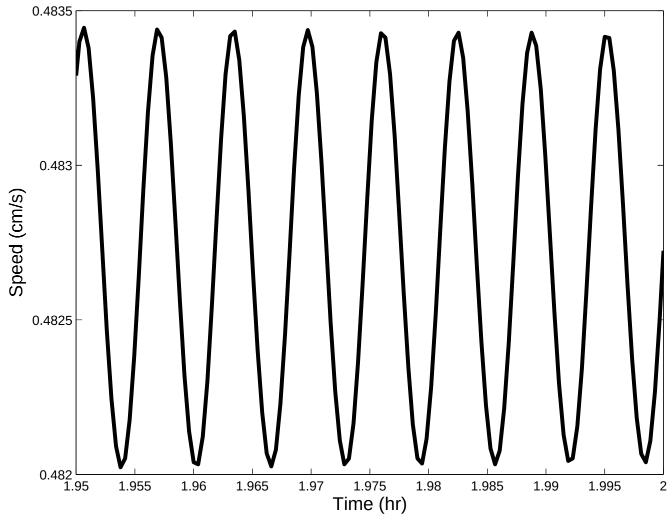

Fig. 6. Flow Stabilization: Speed vs. Time, $K_c = 0.75$

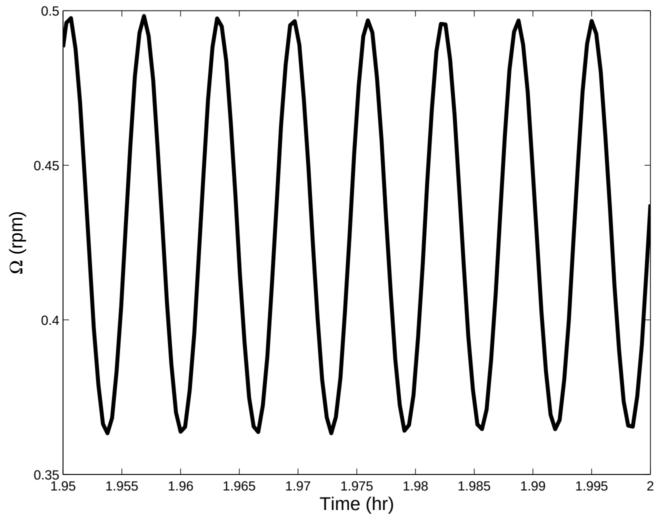

Fig. 8. Flow Stabilization: Ω vs. Time, $K_c = 0.75$

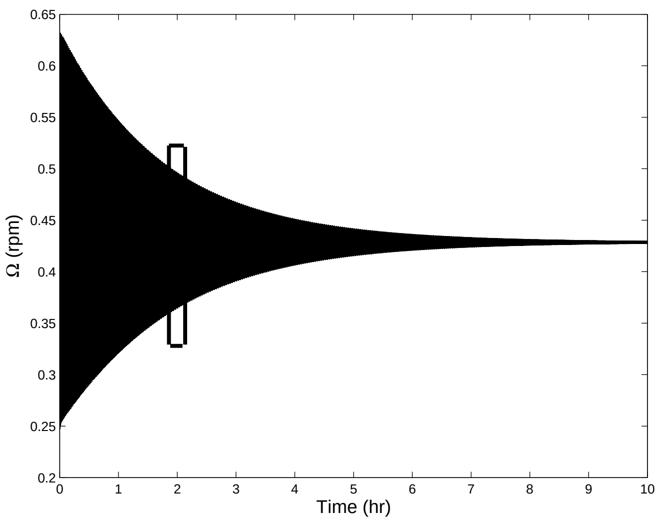

Fig. 7. Flow Stabilization: Ω vs. Time, $K_c = 0.75$

Figure 9 shows the effectiveness of proportional control at attenuating the flow oscillations, as a function of the controller gain, K_c. Controller effectiveness is quantified by a ratio, γ, which is defined as the closed-loop oscillation amplitude divided by open-loop oscillation amplitude. For values of K_c ranging between 0.5 and 0.75, proportional control is effective at stabilization ($\gamma \approx 0$). At values of K_c below this range, rotation has virtually no effect on the flow ($\gamma \approx 1$). At values above this range, rotation appears to exacerbate the instability, causing oscillations of amplitude considerably larger than the open-loop case ($\gamma > 1$).

These simulations demonstrate that feedback control can be used in conjunction with crucible rotation to effectively modify the flow in a crystal growth system for directional solidification. Proportional control is shown to adequately attenuate flow oscillations that arise naturally due to the inherent instability of the system. Issues remain with the practical application of control to the vertical Bridgman system, however. Due to the use of an enclosed crucible, and the need to conduct growth inside a high temperature furnace, it is extremely difficult to measure the states during growth. In practice, flow states could be related to thermocouple measurements, as done by Kim, Witt, and Gatos [1], estimated by eddy sensors [12], or estimated by detailed crystal growth models. Another issue is the speed with which the oscillations are stabilized. The optimal proportional controller required approximately nine hours (or 2000 oscillation periods) to fully attenuate the oscillations. In this work, due to the wide disparity between the crystal growth time scale (hours) and the oscillation time scale (seconds), the furnace was held fixed with respect to time. In practice, however, a control action that requires hours to stabilize the flow is undesirable. We anticipate that a nonlinear, model-based controller would provide a considerably faster response. In addition to improvement of the control algorithm, other types of actuation could be investigated. Our results demonstrate the feasibility of controlling flows at Rayleigh numbers slightly above the critical value, but it remains uncertain whether boundary actuation can effectively suppress oscillations in more strongly nonlinear flows. Other types of actuation that might be feasible for this application include real-time furnace adjustment, or application of lasers (pointwise heating), magnetic fields, or vibration.

V. Concluding Remarks

We have applied proportional controllers to our detailed models of the vertical Bridgman crystal growth process. Control was successful at stabilizing flow oscillations that occur naturally due to the inherent instability of the system. Our results give hope to the idea that feedback process control can be used to prevent the occurrence of compositional striations, thereby improving the quality of crystals grown by the vertical Bridgman method. Our simulations indicate that there is an optimum range for controller gain, within which oscillations can be completely suppressed. Controller gains below this range have little

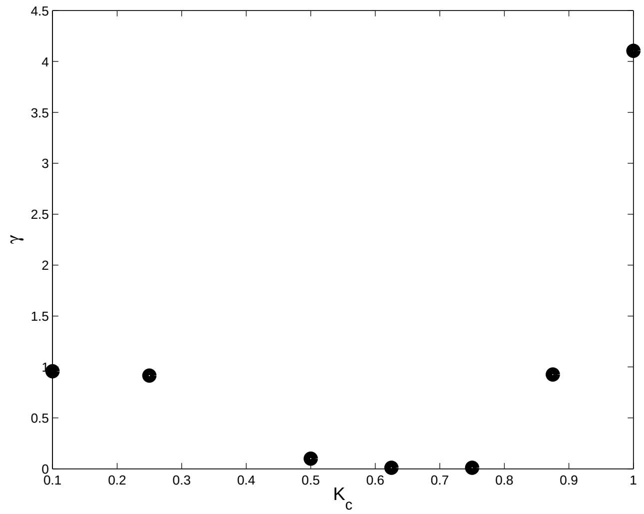

Fig. 9. Effect of Controller Gain on Oscillation Amplitude

affect on the oscillations, and controller gains above this range act to further destabilize the process. Opportunities for future work include the implementation of control algorithms that will accelerate the closed-loop response, and the investigation of other actuation techniques.

Acknowledgments

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