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CONVECTIVE EFFECTS IN FLOAT-ZONE AND CZOCHRALSKI MELTS

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The hydrodynamics of crystal-growth melts is a relatively new research area. Numerical modeling of these processes is necessary, considering the hostile environment from the standpoint of laboratory experimentation. The work discussed herein is in two parts: numerical simulations are being conducted of the flow in a Czochralski melt and also of that in a float zone. In addition, for the float-zone case, energy stability theory will be used to determine stability bounds for the onset of oscillatory thermo-capillary flow.

Convective effects in crystal-growth melts arise from a variety of mechanisms. Temperature gradients both in the direction of gravity and normal to it give rise to convection due to buoyancy effects. Rotation of the crucible and/or crystal causes a forced convection which may augment or oppose the buoyancy-driven flow. Finally, thermo-capillary forces (due to the variation of surface tension with temperature) drive surface motions which in turn generate convection in the bulk fluid. All of these mechanisms are present in either Czochralski or float-zone growth.

The research objective of the Czochralski modeling is to develop an accurate numerical simulation of the flow in a Czochralski silicon melt and to investigate the effects of various parameters on the flow properties. Like some earlier investigations, we intend to simulate the effects of buoyancy, forced, and thermo-capillary convection, including unsteady effects. Unlike earlier work, the ultimate aim is to be able to include the effects of a variable free surface and freezing interface and, possibly incorporate non-axisymmetric effects.

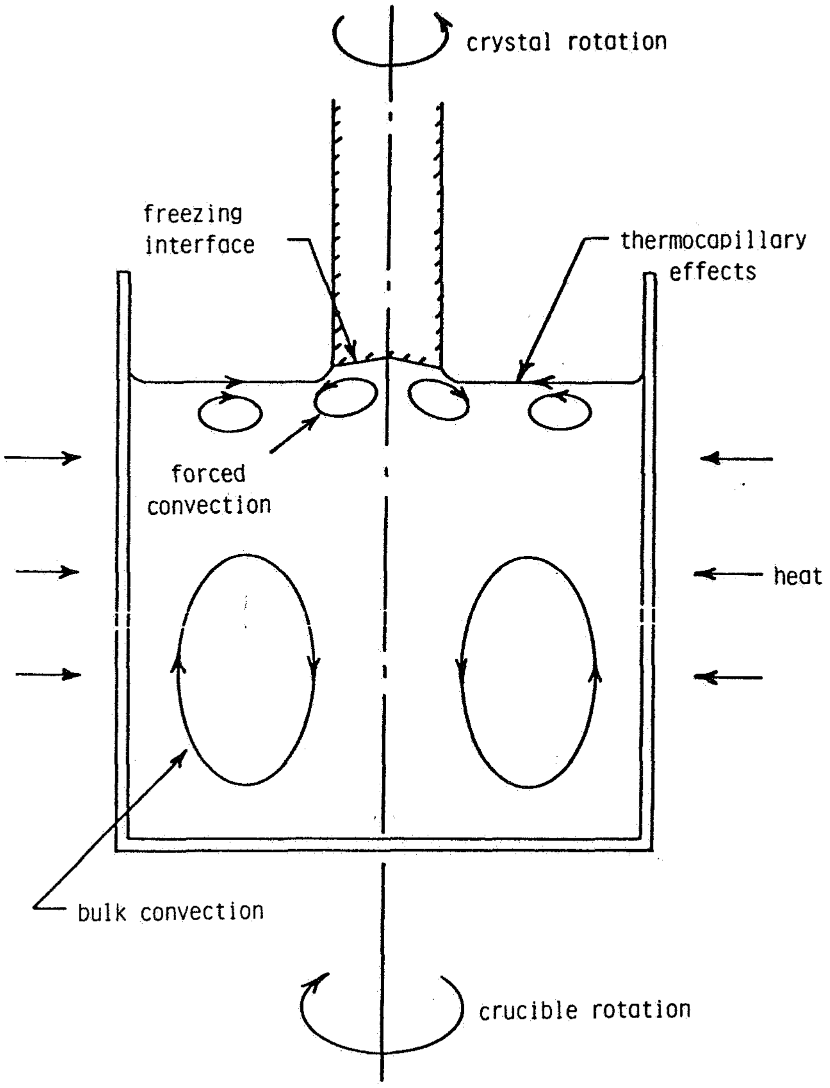
Model experiments by Preisser, Schwabe and Scharmann and also Chun have demonstrated the importance of thermo-capillary convection on the float-zone process. In particular, the work of the former group has concluded that it is an instability of thermo-capillary convection which leads to the onset of oscillatory convection which may be responsible for striations observed in some crystals. This phenomenon will be especially important in applications of the float-zone process in a microgravity environment.

Computations of float-zone basic states will concentrate on three model problems. The first of these will neglect free-surface deformation and consider flat melting and freezing interfaces. The second will relax the restriction on free-surface deflection, but maintain flat crystal/melt interfaces. Finally, the third model problem will attempt to compute basic states for which these interfaces are determined as part of the problem.

A stability analysis will be performed on each of these basic states using energy stability theory. Energy theory yields a sufficient condition for stability, i.e., a "critical" value of a dimensionless parameter below which disturbance growth will not take place. For some types of instability mechanisms, the sufficient condition for stability provided by energy theory is quite conservative, i.e., small. For instabilities driven by surface tension, however, there is reason to believe that this will not be the case. Hopefully, the results of the study will provide results which will allow the growth of striation-free crystals by the float-zone process.

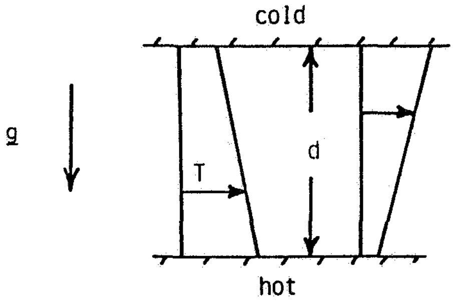
The computations will be performed for each of the basic states discussed above under conditions of variable gravity to examine the possibilities for space processing by this technique. The consideration of the proposed basic states will allow comparisons with previous analytical work by Davis and his coworkers and the model experiments of Preisser et al. mentioned above. These will assist greatly in the development and verification of the model.

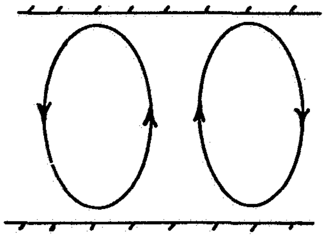
Czochralski Process



Convection Mechanisms in Cz Crystal Growth

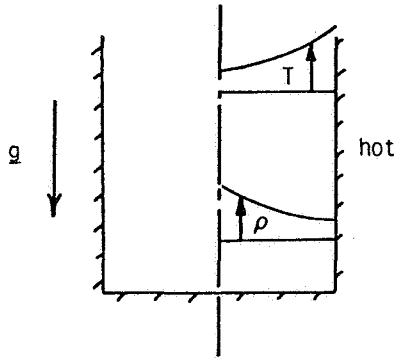
I. Buoyancy

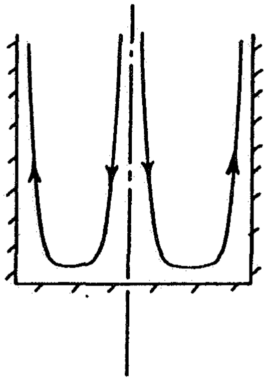




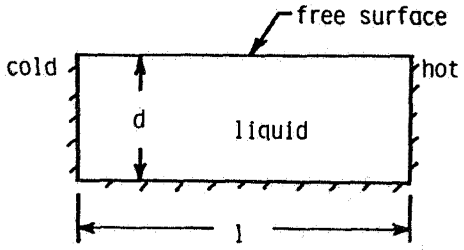
Rayleigh number $R = \frac{\alpha \, \Delta T \, g \, d^3}{\kappa \, \upsilon}$

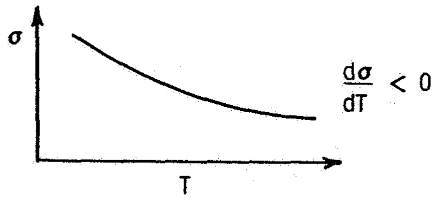
Prandtl number $Pr = \frac{\upsilon}{\kappa}$

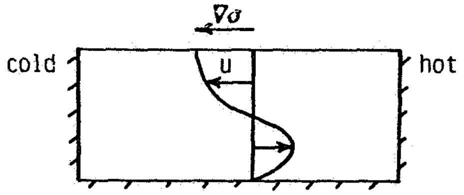




Thermocapillary Effects







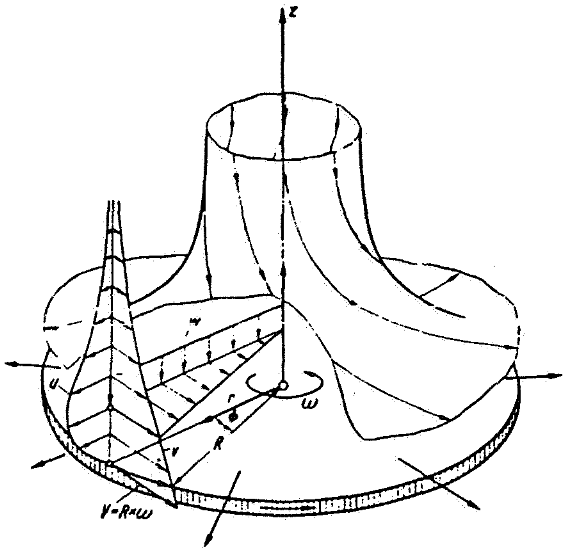
Aspect ratio $A = d/l$

Marangoni number $M = \frac{-d\sigma/dT \; A \, \Delta T \; d}{\mu \kappa}$

Capillary number $C = \frac{-d\sigma/dT \; A \, \Delta T}{\sigma}$

Forced Convection

Flow "above" a rotating disc:



Reynolds number $\mathrm{Re} = \frac{\Omega R^2}{\upsilon}$

Research Objective

Numerical simulation of the flow and temperature fields occurring during Czochralski growth of silicon.

Investigation of the influence of various parameters (e.g., rotation, aspect ratio) on the results.

Ultimate Computational Abilities

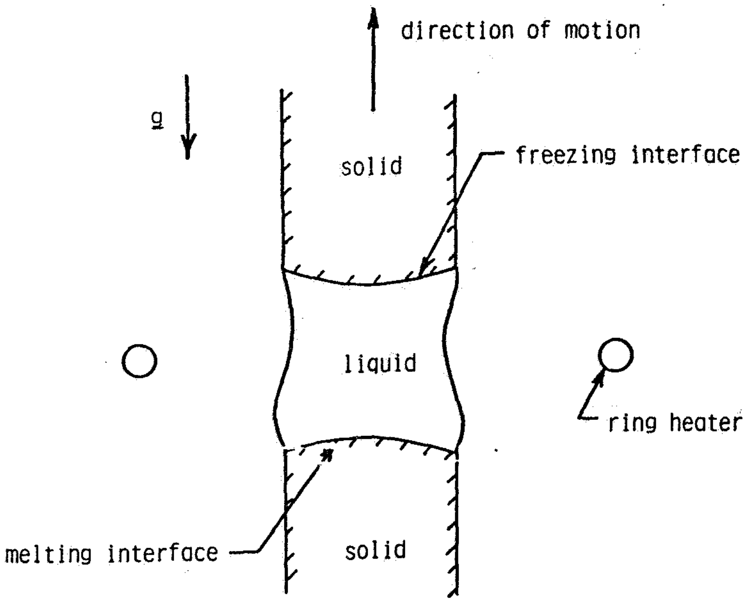
Effects of buoyancy, forced, and thermocapillary-induced convection.

Time-dependent simulation.

Variable free-surface and freezing interface.

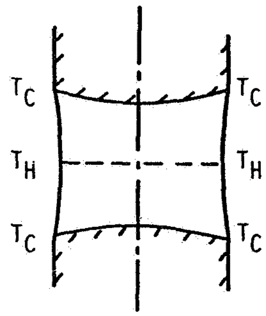
Non-axisymmetric simulation ? ?

Float-Zone Process

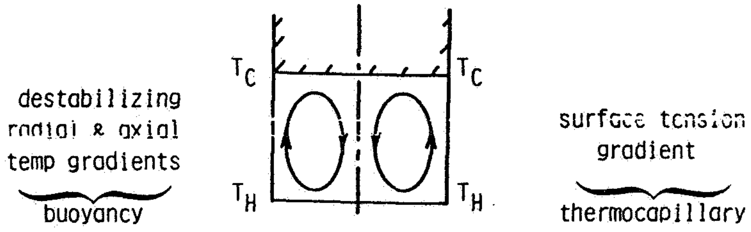


Advantage: containerless process

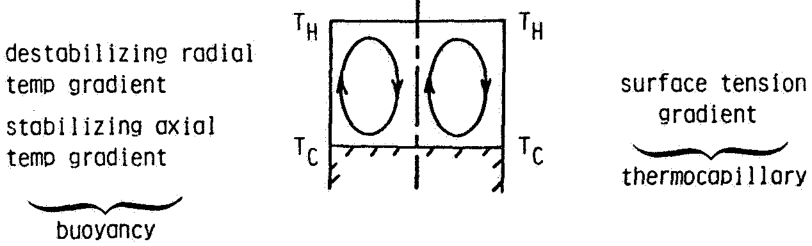
Float zone



Top half:

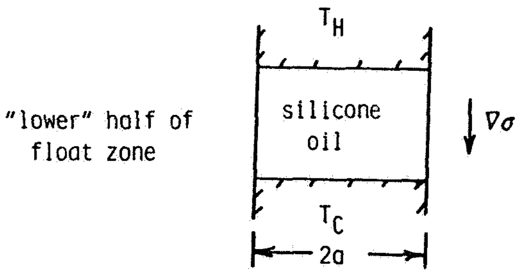


Bottom half:



Model Experiments

Preisser, Schwabe & Scharmann have performed model experiments in which the dominant convection mechanism is thermocapillarity:

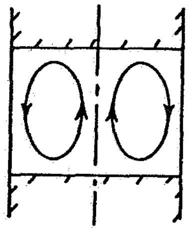


Relevant dimensionless parameter:

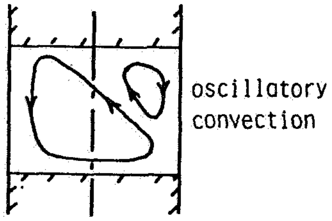
Marangoni number $$M = \frac{\rho c_p |d\sigma/dT| \cdot |dT/dz| a^2}{\mu k}$$

Results:

$M < O(10^4)$



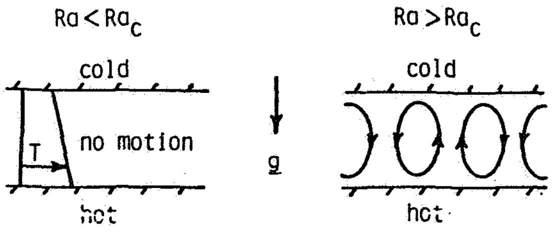
$M > O(10^4)$



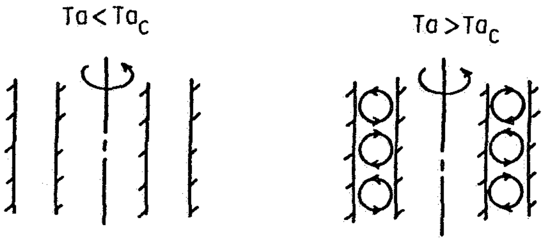
Hydrodynamic Stability

Like other dynamical systems, under certain situations, some fluid flows can be unstable to the inevitable disturbances which are present. In some cases, the instability of a given basic state results in the establishment of another laminar state:

Rayleigh/Benard convection



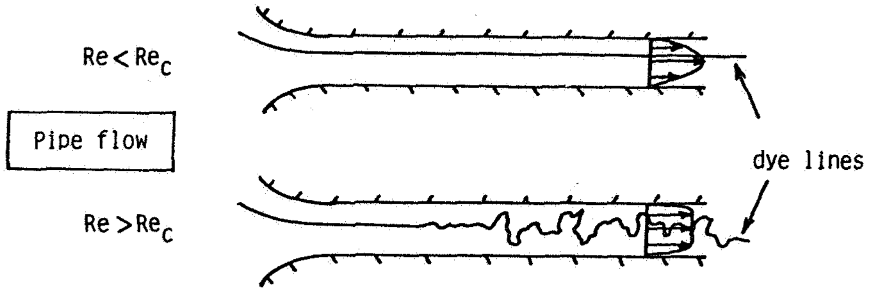
Taylor/Couette flow



pure swirl

Taylor vortex flow

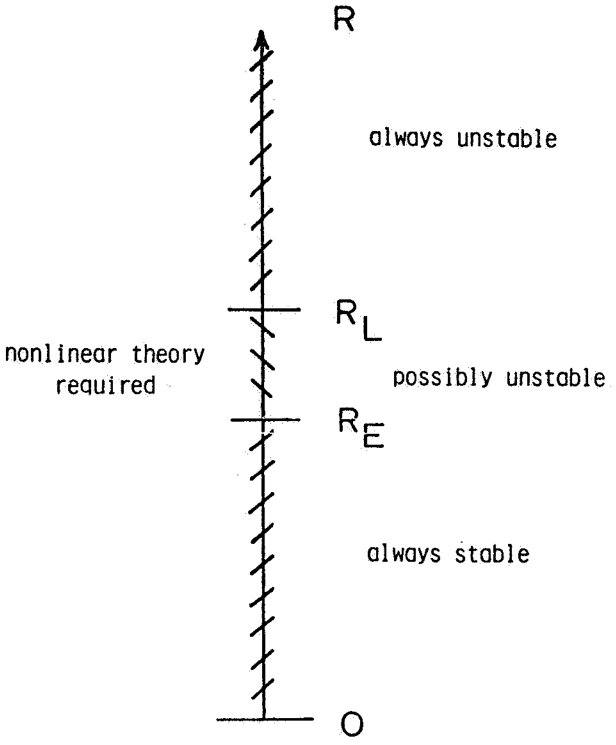
In other cases, the instability can lead directly to a chaotic, turbulent state:



In each case, there is a relevant dimensionless parameter; when it exceeds a "critical" value, the flow becomes unstable.

The goal of the present research is to examine the stability of thermocapillary convection in models of the float-zone crystal growth process in terrestrial and microgravity environments.

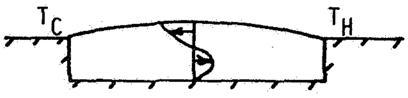
Stability Criteria



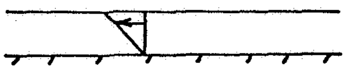
Previous Related Work (Linear Stability Theory)

Sen & Davis, Smith & Davis

2-D slot

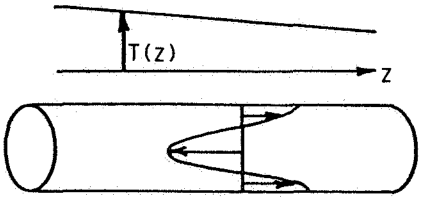


2-D layer with
surface shear stress



Instability mechanisms studied:

- classical shear flow
- surface waves
- convective instabilities
  - rolls
  - "hydrothermal waves"



basic state requiring return flow

The preferred instability was found to be a hydrothermal wave.

- Similar azimuthal dependence as the oscillatory convection seen in model float-zone experiments
- Much lower critical Marangoni number than that observed experimentally.

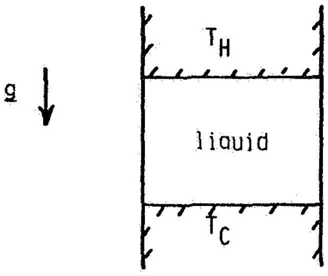
  Reasons (speculated):

  finite length is a stabilizing feature.

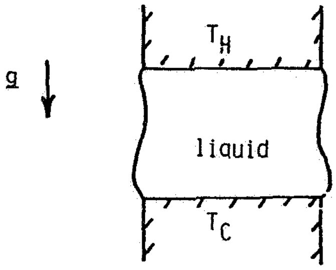
  buoyancy forces in the model half-zone experiment are stabilizing.

Problem Sequence

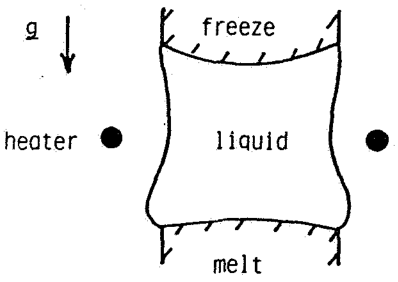
Features



- model half zone
- specified planar endwalls
- non-deformable free-surface
- variable body force



- model half zone
- specified planar endwalls
- deformable free-surface
- variable body force



- float-zone configuration
- unspecified melt/freeze interfaces
- deformable free-surface
- variable body force

DISCUSSION

MATLOCK: In silicon float-zone crystal growth, the heating is always done with radio frequencies, where there is significant RF levitation. Have you considered the possibilities of what that will do to the validity or invalidity of the directions that you are going?

NEITZEL: We have considered the possibilities but there is also some work being done on focused light melting, so we feel that the exclusion of the RF effects might not be unrealistic for that type of a case.

MATLOCK: Do you have any general thoughts about what the RF situation would do to what you are doing?

NEITZEL: No, not really. It's an additional complication that adds to an already very complicated problem.

As an aside, I would like to emphasize that there is a great need for these hydrodynamic simulation experiments because one can't make fluid dynamical measurements in molten silicon. It's a very hostile environment. It's optically opaque. If we have a model experiment in which certain other relevant dimensionless parameters might be matched, then that allows us to have something to compute analytically or numerically. And then, if we can verify a numerical model against that model experiment, that gives us some justification for extrapolating to other values or parameters that might be more appropriate to Czochralski growth.

KIM: Thermal conductivity is completely different, but you may simulate fluid dynamics like a Marangoni flow. If you include axial non-symmetric flow, can you still use a 2-D simulation or do you have to go to a 3-D simulation?

NEITZEL: One does have to make a three-dimensional simulation. The equations are not separable. You cannot just take an $e^{im\Theta}$-type dependence because of the fact that you have non-linearities. You have to do a three-dimensional simulation, but there have been advances recently in numerical fluid dynamics and in the use of structural methods that seem to hold great promise for doing this kind of computation. So it's not out of the realm of possibility.

ELWELL: Do you have a good number for $d\sigma/dT$ for silicon?

NEITZEL: No, there are several numbers in the literature, but I don't think there is any universal agreement as to what the $d\sigma/dT$ is. I'm not sure there is universal agreement as to what σ is.

KALEJS: I believe there is a recent measurement by Hardy that Bob Brown mentioned at NBS that is about 0.2 or so. It is found in an internal NBS publication.

C-2

WITT: We tried elements of what you have tried to do in a reduced-gravity environment. We did indium antimonide and the theoretical predictions were that the driving forces for Marangoni flow exceed by a factor of 10 each of the driving forces for buoyancy-driven convection. It means that the phenomenon is strictly and exclusively due to Marangoni-type convection. We did the experiment in space and there was zero evidence for any Marangoni-type flow. I'm not saying that the theory is wrong. I am throwing an enormous amount of doubt on the available $d\gamma/dT$ data, because if you look at the sensitivity of the surface tension to contamination, it's fierce. Not only that, we also have absolutely no numbers for the gradients in the vicinity of the growth interface itself. So you are confronted with non-availability of thermo-physical data and an absolute lack of quantitative verification of thermal gradients and then you have to attempt to interpret the data. Finally, I wanted to say that in your system you have radial thermal gradients and then regardless of your stabilizing axial gradient, you will have serious convection.

NEITZEL: The magnitude of this convection is probably swamped by thermocapillary effects.

WITT: If you do it in confinement then your surface-tension-driven convection falls down to almost nothing. In the Bridgman configurations, we find that the momentum boundary layer is in the micron range stabilized with minimized radial gradients. It cannot get much less.

NEITZEL: You are not talking about a model experiment with silicon oil. You are talking about a crystal-growth experiment with radial gradients.

WITT: No, you are now making a dangerous conclusion, by drawing from one system conclusions to another. The surface-tension sensitivity in silicon oil cannot be compared to that of metallic melts. Therefore, the conclusions to be drawn are very, very dangerous, because you have no surface-tension data. I don't think anybody will question the soundness of your argument that there are surface-tension forces and they give rise to convection. However, the magnitude of the driving forces is in many instances in competition with other driving forces. Then the question is what becomes the primary mode. That's where I think we have a problem.

NEITZEL: That's right, but all we can do with our modeling is to try to predict those types of instabilities that occur where we know what's going on. That silicon oil experiment is a very good example of that. The data are very well known there. The material properties are well known, and if we can succeed in giving a stability boundary for that type of flow, then we have some confidence that maybe we could predict, if given the right material properties, what would happen in a float-zone silicon melt.

DYER: Is the wall pumping in the Czochralski arrangement included in your model? Being of a larger area and the fact that they are rotating, possibly counter-rotating, this would seem to be an overwhelming effect compared with the small size of the crystal.

NEITZEL: What's really important to the crystal-growth process is what's going on in the vicinity of the freezing interface. Whereas that may be a very small effect in the overall bulk convective motion, it might be a very important effect with regard to the transport of scale of properties, oxygen, heat and whatsoever in the vicinity of the freezing interface. The side walls don't really do any pumping in the sense that the end walls do pumping and that the crystal-freezing interface does pumping.