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INTERACTIONS BETWEEN SOLIDIFICATION AND COMPOSITIONAL CONVECTION IN MUSHY LAYERS

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INTRODUCTION

Mushy layers are ubiquitous during the solidification of alloys. They are regions of mixed phase wherein solid crystals are bathed in the melt from which they grew. The matrix of crystals forms a porous medium through which the melt can flow, driven either by external forces or by its own buoyancy in a gravitational field. Buoyancy-driven convection of the melt depends both on temperature gradients, which are necessary for solidification, and on compositional gradients, which are generated as certain components of the alloy are preferentially incorporated in the solid phase and the remaining components are expelled into the melt. In fully liquid regions, the combined action of temperature and concentration on the density of the liquid can cause various forms of double-diffusive convection. However, in the interior of mushy regions the temperature and concentration are thermodynamically coupled so only single-diffusive convection can occur. Typically, the effect of composition on the buoyancy of the melt is much greater than the effect of temperature, and thus convection in mushy layers is driven primarily by the compositional gradients within them.

It has long been recognized that compositional convection within mushy layers can become focused into narrow channels, called 'chimneys', which are identified as 'freckles' in completely solidified castings. Freckles destroy the structural and compositional uniformity of a casting, in most cases rendering the casting useless. There has consequently been much interest in determining the conditions under which chimneys will form during casting and in determining ways in which their formation might be suppressed.

Chimneys and the associated compositional convection have been observed in many laboratory experiments in which aqueous solutions of ammonium chloride were cooled and solidified from below [1–3], and freckles that are visually similar to the ammonium-chloride chimneys have been observed in experimental castings of lead–tin alloys solidified from below [4]. In both these systems, the light component of the alloy is rejected during solidification so that the interstitial melt in the mushy layer is less dense than the un-solidified melt above it and can therefore rise convectively out of the mushy layer.

The rising interstitial liquid is relatively dilute, having come from colder regions of the mushy layer, where the liquidus concentration is lower, and can dissolve the crystal matrix through which it flows. This is the fundamental process by which chimneys are formed. It is a nonlinear process that requires the convective velocities to be sufficiently large, so fully fledged chimneys might be avoided by means that weaken the flow. Better still would be to prevent convection altogether, since even weak convection will cause lateral, compositional inhomogeneities in castings. This report outlines three studies that examine the onset of convection within mushy layers.

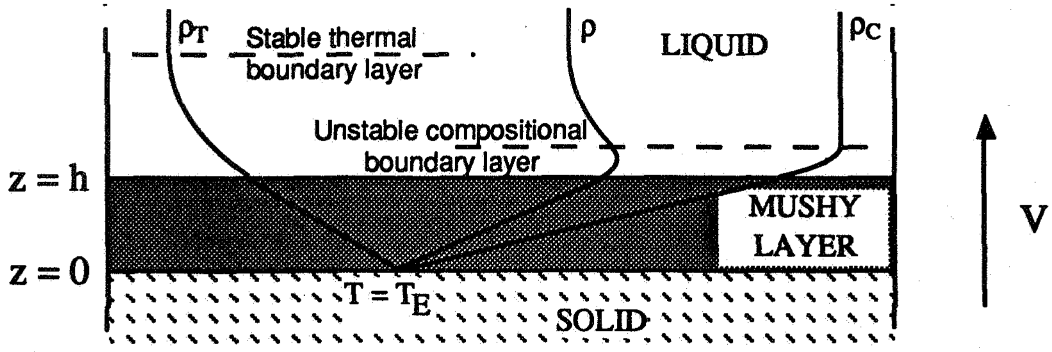

Figure 1. A schematic diagram showing the steady upwards solidification of an alloy at speed V. The steady density field ρ is indicated. It is the resultant of the density fields ρ_T due to the temperature gradient and ρ_C due to the compositional gradient.

LINEAR STABILITY ANALYSIS

A convenient system for mathematical analysis is one in which the alloy is solidifying at a constant rate V, as shown in figure 1. In a frame of reference moving at the solidification rate, governing equations for the mushy layer are

$$\left(\frac{\partial \theta}{\partial t} - \frac{\partial \theta}{\partial z}\right) + \mathbf{U}.\nabla\theta = \nabla^2\theta + S\left(\frac{\partial \phi}{\partial t} - \frac{\partial \phi}{\partial z}\right), \tag{1}$$

$$(1-\phi)\left(\frac{\partial \theta}{\partial t} - \frac{\partial \theta}{\partial z}\right) + \mathbf{U}.\nabla\theta = (\theta - \mathcal{C})\left(\frac{\partial \phi}{\partial t} - \frac{\partial \phi}{\partial z}\right), \tag{2}$$

$$\nabla.\mathbf{U} = 0, \tag{3}$$

$$\frac{\mathbf{U}}{\Pi(\phi)} = -R_m(\nabla p + \theta\hat{\mathbf{z}}), \tag{4}$$

where

$$\theta = \frac{T - T_L(C_0)}{T_L(C_0) - T_E} = \frac{C - C_0}{C_0 - C_E}, \tag{5a, b}$$

represents both the temperature T and the concentration C in the mushy layer, ϕ is the solid fraction, and $\mathbf{U}$ is the fluid velocity. In these equations, lengths have been scaled with the thermal diffusion length $H = \kappa/V$, time with κ/V^2 and fluid velocities with V, where κ is the thermal diffusivity. These equations are augmented by the buoyancy-driven Navier-Stokes equations and diffusion-advection equations for heat and solute in the fully liquid region, as well as various boundary and interfacial conditions [5].

The important dimensionless parameters governing the evolution of mushy layers are the Stefan number S and a compositonal ratio $\mathcal{C}$, gvien by

$$S = \frac{L}{c(T_L(C_0) - T_E)} \quad \text{and} \quad \mathcal{C} = \frac{C_s - C_0}{C_0 - C_E}, \tag{6a, b}$$

where c and L are the specific heat and latent heat respectively,and a parameter θ_∞, which can be interpreted either as the dimensionless far-field temperature in the liquid or as the temperature gradient at the mush–liquid interface in the basic, steady state. Compositonal convection is determined principally by the Rayleigh numbers

$$R_m = \frac{\beta\,(C_0 - C_E)\,g\,\Pi^*\,H}{\kappa\,\nu}, \tag{7}$$

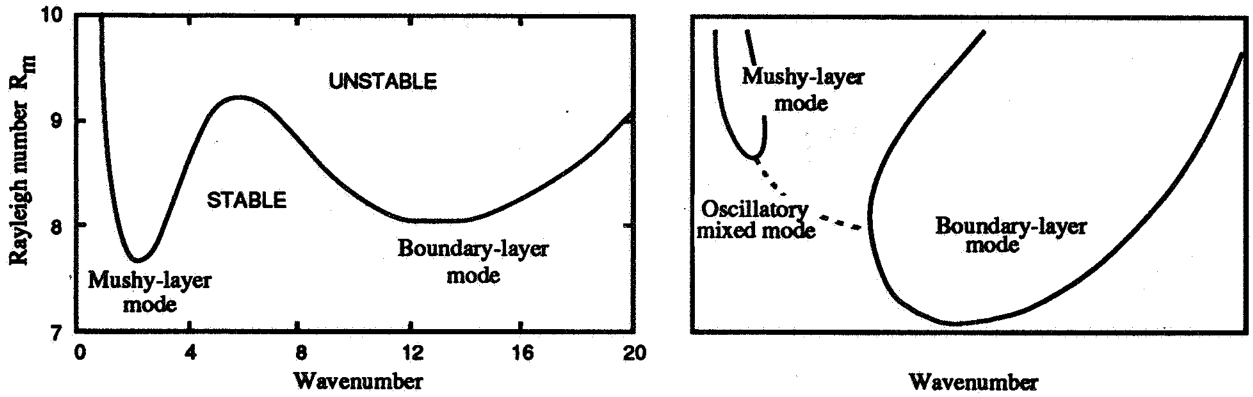

Figure 2. (a) A marginal stability curve for convection within and above a mushy layer [6] when the stabilizing thermal buoyancy is neglected. Parameter values were chosen so that the two modes of instability would occur at about the same Rayleigh number. Either mode can, in principle, be the most unstable. (b) A sketch of a typical marginal stability curve once thermal buoyancy is included in the analysis [8]. Parameter values are chosen such that the boundary-layer mode occurs well before the mushy-layer mode, which is the situation in most experiments.

which measures the strength of buoyancy relative to the resistance to flow in the mushy layer and $R_{\delta C} = \epsilon^3 \mathcal{H} R_m$, which is a compositional Rayleigh number based on the buoyancy and dimensions of a compositional boundary layer above and adjacent to the mush–liquid interface. The physical parameters entering these expressions are the solutal expansion coefficient β, the acceleration due to gravity g, the kinematic viscosity of the liquid ν and a characteristic permeability of the mushy layer Π^*. The dimensionless parameter $\epsilon = D/\kappa$, where D is the compositional diffusivity, is typically very small, while the mobility ratio $\mathcal{H} = H^2/\Pi^*$ is typically very large.

The governing equations have a steady solution with no flow [5–7]. The density field associated with this steady state is illustrated in figure 1. Most of the unstable buoyancy occurs within the mushy layer, where the fluid is relatively immobile. There is a thin, unstable compositional boundary layer above the mushy layer, where the fluid is relatively mobile. A linear stability analysis [6] shows that these two unstable regions give rise to two modes of compositonal convection: a mushy-layer mode and a boundary-layer mode (figure 2a). Either mode can in principle be the most unstable, though experimental evidence suggests that thé boundary-layer mode usually occurs first. The linear-stability analysis has recently been extended [8] to include the stabilizing thermal buoyancy in the liquid region. The thermal buoyancy can cause the two steady modes to disconnect via an oscillatory mode of convection (figure 2b) that is presumably related to internal gravity waves in the stably-stratified liquid region. The stability of a transient basic state in which the mushy layer is growing away from a cooled boundary has also been analysed using a quasi-static approximation [9].

Influenced by experiments [10] in which chimneys were initiated by sucking fluid from the boundary layer with a pipette and by the experimental observation that convection emanating from the boundary layer usually preceeds chimneys, some argue that chimneys are caused by convection in the liquid region (i.e. the boundary-layer mode) eroding the layer from above. It is more plausible, in my opinion, that chimneys are formed by the mushy-layer mode of convection eroding the layer from within. However, since the boundary-layer mode is seen to occur first in experiments, it is important to investigate what influence this mode has on the mushy layer and to ask whether the stability analysis of a quiescent state has any relevance to chimney-forming convection in the mushy layer.

The boundary-layer mode of convection has characteristic length scales that are much smaller than the overall dimensions of the mushy layer and the characteristic length scale of the mushy-layer mode. Emms & Fowler [9] exploited this fact and homogenized a model of fully developed convection above the mushy layer to determine spatially averaged boundary conditions at the mush–liquid interface. One of the results of their analysis is that pre-existing convection above the mushy layer has little influence on the onset of convection within the mushy layer. However, as described in the next section, the story is different once non-equilibrium effects are taken into account.

INTERFACIAL UNDERCOOLING

The temperature of solidifying phase boundaries is below the equilibrium freezing (liquidus) temperature. Further undercooling at a mush–liquid interface is caused by the curvature of the dendrite tips and constitutional effects associated with the compositional boundary layer. Usually such departures from equilibrium are small and can safely be neglected in macroscopic models of alloy solidification. However, it has been shown that interfacial undercooling can have large, global consequences when coupled with convection of the liquid region [11].

Huppert & Hallworth [12] conducted experiments in which they observed that the occurence of chimneys in ammonium-chloride mushy layers could be delayed by the addition of small quantites of copper sulphate to the solution. In an attempt to explain these observations, Worster & Kerr [13] measured the undercooling at the mush–liquid interface in similar experiments. They found that the level of undercooling increased with the rate of solidification and with the level of contamination by copper sulphate. They incorporated this empirical information into a macroscopic model of the solidifying system in which the liquid region is assumed to be convecting vigorously (the boundary-layer mode) while the mushy layer is stagnant.

In accord with the experimental results [12], the mathematical model [13] predicts that, as the level of interfacial undercooling increases, the mushy layer grows more slowly, the solid fraction of the mushy layer increases and the composition of the liquid region evolves more rapidly. These are all consequences of the fact that the strength of compositional convection in the liquid region increases as the level of undercooling increases. The resultant increase in solute flux causes the solid fraction to increase and the liquid region to evolve more rapidly. The increase in heat flux and the increase in release of latent heat cause the mushy layer to grow more slowly. These three effects all combine to cause the Rayleigh number associated with the mushy layer to increase more slowly, and can cause the Rayleigh number eventually to decrease (figure 3a). The appropriate Rayleigh number is given in (7), with H being the height of the mushy layer, $C_0 - C_E$ being replaced by the compositional difference across the mushy layer, and with Π^* being evaluated in terms of the mean solid fraction according to an expression for a regular array of cylinders [3].

The height of the mushy layer when the Rayleigh number is predicted to have reached various fixed values is shown in figure 3b. On the same figure, data [12] of the heights of the mushy layer when chimneys were first observed are shown. Comparison between the data and the theoretical predictions lends some support to the idea that chimneys occur once a critical Rayleigh number (*for the mushy layer mode*) is exceeded and that compositional convection in the liquid region, in concert with interfacial undercooling, can significantly alter the propensity of the mushy layer to form chimneys.

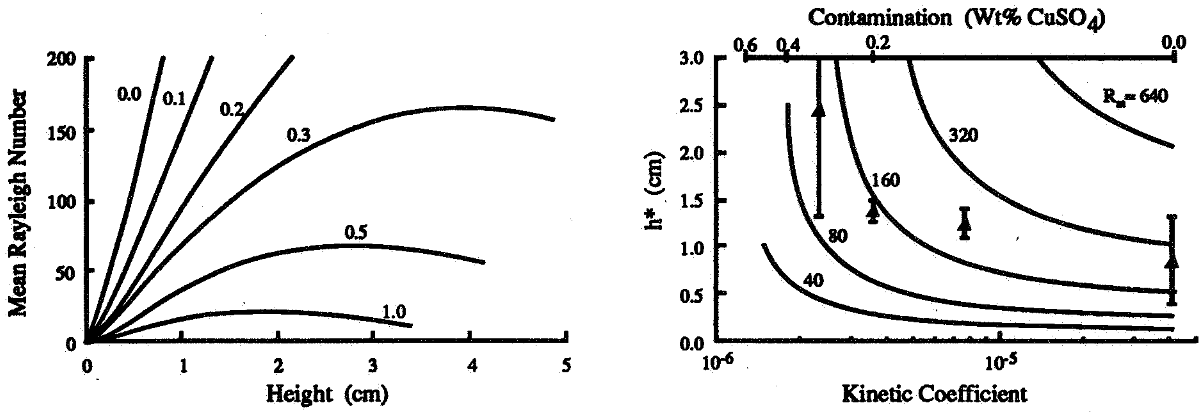

Figure 3. (a) Predicted values of the mean Rayleigh number of the mushy layer as functions of the depth of the layer, as it evolves while vigorous compositional convection driven by interfacial undercooling occurs in the liquid region. The numbers on the curves correspond to different levels of undercooling. The Rayleigh number reaches a maximum, which decreases as the level of undercooling increases. (b) The critical height at which given values of the Rayleigh number (indicated by the numbers on the curves) are first reached as function of the kinetic coefficient, which measures the level of undercooling.

NONLINEAR BIFURCATIONS

Linear stability analyses determine the critical Rayleigh number R_c above which the solidifying system is unstable to infinitesimal disturbances. However, it has long been suspected that the bifurcation from quiescent to convecting states is sub-critical, i.e. that steady convecting states (perhaps giving rise to chimneys) exist at Rayleigh numbers less than R_c. In this case, linear stability analyses may give inadequate indication of the conditions required to avoid chimney-forming convection.

This suspicion was confirmed using a small-amplitude analysis of the mushy-layer mode of convection [14]. Use was made of a 'near-eutectic' approximation of the governing equations [15] which is achieved in the asymptotic limit $\mathcal{C} \to \infty$, $\theta_\infty \to \infty$, with $\mathcal{C}/\theta_\infty = O(1)$. Effects of latent heat can be additionally retained in the analysis if the distinguished limit $\mathcal{S}/\mathcal{C} = O(1)$ is taken [9]. This limit reduces the equations for a mushy layer to those of a non-reacting porous medium of constant permeability. Effects fundamental to the character of mushy layers, namely the coupling between the temperature and concentration, and the dependence of permeability on the solid fraction, can then be introduced as perturbations to the simpler system.

It was discovered [14] that the bifurcation to convective rolls could be either super-critical (as shown in figure 4a) or sub-critical depending on the magnitude of $d\Pi/d\phi$, and that the bifurcation to convection with a hexagonal planform is trans-critical, with the backward branch corresponding to upflow in the centres of the hexagons. The form of the trans-criticality that was found is due to nonlinear perturbations to the permeability of the mushy layer.

Trans-critical hexagonal convection may also result from asymmetries of the basic-state density field and solid fraction. These effects may cause the backward branch of the trans-critical bifurcation to correspond to downflow in the centres of the hexagons (figure 4b), in agreement with experimental observations [16]. If this is the case, then a judicious choice of control parameters may eliminate the backwards branch altogether and render the system globally more stable to chimney-forming convection. An analysis of this extended system, including the determination of

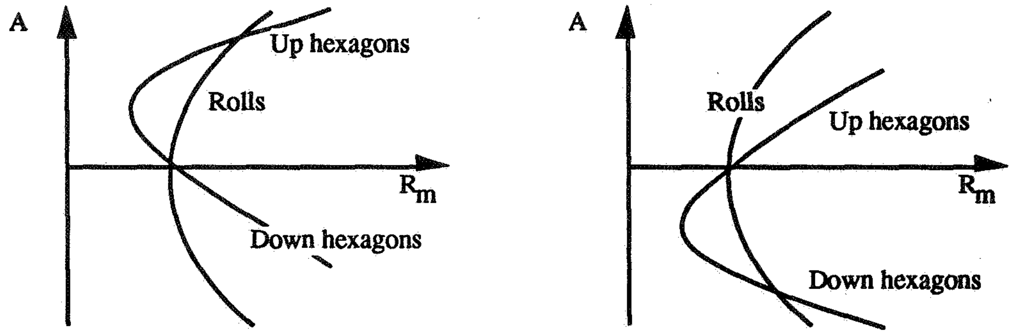

Figure 4. Schematic, nonlinear bifurcation diagrams showing the amplitude of steady, convecting states as functions of the Rayleigh number. (a) Trans-critical bifurcation to hexagons, with the backward branch corresponding to upflow in their centres, has been found by [14] as a consequence of nonlinear perturbations to the permeability of the mushy layer. (b) Asymmetries in the quiescent state may lead to the backward branch corresponding to downflow in the centres of the hexagons.

the stability characteristics of the nonlinear solutions is currently being undertaken.

CONCLUSIONS

Linear stability analyses indicate that there are two modes of compositional convection that can occur during the solidification of a binary alloy from below: a boundary-layer mode of very short wavelength compared with the depth of the mushy layer; and a mushy-layer mode that is most likely to be responsible for the formation of chimneys. Vigorous convection in the liquid region (originating as the boundary-layer mode) can couple with interfacial undercooling at the mush–liquid interface and reduce the liklihood of chimneys forming in the mushy layer. A nonlinear analysis of convection interior to a mushy layer, currently under way, promises to yield conditions under which the solidifying system is globally more stable to the formation of chimneys.

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