CONVECTIVE INSTABILITIES DURING SOLIDIFICATION OF A MUSHY LAYER

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<u>Summary</u> The convective instabilities of binary alloys solidified from a bottom boundary are analyzed under linear stability theory. By using a similarity variable, the time-dependent disturbance equations for the liquid and mushy layers are transformed to the self-similar stability equations. The critical conditions at the onset of convection in the mushy layer are found numerically for the solidification of aqueous ammonium chloride solution.

INTRODUCTION

Convective instability during solidification causes freckles in a resultant solid and often controls the quality of solid products in the processes of crystal growth and castings of metallic alloys. In binary solidification, a mushy layer of dendritic crystals is often formed by constitutional supercooling. When a binary alloy is solidified in a gravitational field, an unstable density profile induced by concentration and temperature gradient may occur buoyancy-driven convection in the mushy layer.

In the present study, the onset of convective instabilities in the mushy layer during time-dependent solidification of binary alloys is analyzed based on propagation theory we have developed. By using a similarity variable, the time-dependent disturbance equations for the liquid and mushy layers are transformed to the self-similar stability equations. The present study suggests the critical conditions to mark the convective motion in the liquid and mushy layers of the solidification system in which the mushy-layer thickness and the density profile vary with time.

The system considered here is shown in Fig. 1. The supereutectic melt is initially quiescent at a constant temperature T_{∞} and a constant solute concentration C_{∞} . For time $t \geq 0$ the bottom boundary of the melt is supercooled at a constant temperature, which is lower than the eutectic temperature $T_{\rm E}$. The mushy layer grows above a eutectic solid layer, and compositional convection of light residual liquid may be induced. It is assumed that the mush-solid interface is at the eutectic concentration $C_{\rm E}$ and the solid-layer thickness at the bottom is very small. The position of the mush-liquid interface $H = 2\lambda \sqrt{\kappa t}$ is moving upward, where λ is the phase-change rate and κ is the thermal diffusivity.

SELF-SIMILAR STABILITY EQUATIONS

The governing equations for convection during solidification of binary alloys are described well by Worster [1] and Emms and Fowler [2]. In the present study, the non-dimensional time-dependent disturbance equations are transformed to functions of a similarity variable ζ (= $z/h = z/(2\lambda\sqrt{\tau})$), where h is the mushy-layer thickness scaled by an arbitrary length L, and τ is the time scaled by L^2/κ . The self-similar stability equations in the mushy layer are given in the previous work [3]:

$$\begin{split} &(\overline{\mathbf{D}}^2 + 2\lambda^2 \zeta \overline{\mathbf{D}} - a^{*2})\theta_m^* = R_m^* w_m^* \overline{\mathbf{D}}\theta_{m0} - 2\lambda^2 St \zeta \overline{\mathbf{D}} \chi^*, \\ & 2\lambda^2 \zeta [\chi^* \overline{\mathbf{D}}\theta_{m0} + \chi_0 \overline{\mathbf{D}}\theta_m^* + (\theta_{m0} - \gamma) \overline{\mathbf{D}} \chi^* + \theta_m^* \overline{\mathbf{D}} \chi_0] = R_m^* w_m^* \overline{\mathbf{D}}\theta_{m0}, \\ &(\overline{\mathbf{D}}^2 - a^{*2})w_m^* = a^{*2}\theta_m^*, \end{split}$$

where $\overline{D} = d/d\zeta$ and a^* is the wave number. The amplitude of velocity disturbance w_m^* has the scale of $\kappa\Pi/L^3$, where Π is the permeability of the mushy layer. In the liquid layer, the amplitude of velocity disturbance is rescaled by a non-dimensional time-dependent scaling factor h^2 (= $4\lambda^2\tau$) based on propagation

theory, and w^* has the scale of $\kappa H^2/L^3$. The self-similar stability equations in the liquid layer are given by

$$\begin{split} &(\overline{D}^{2} + 2\lambda^{2}\zeta\overline{D} - a^{*2})\theta^{*} = R_{T}^{*}w^{*}\overline{D}\theta_{0},\\ &(Le\overline{D}^{2} + 2\lambda^{2}\zeta\overline{D} - Lea^{*2})c^{*} = R_{S}^{*}w^{*}\overline{D}c_{0},\\ &[(\overline{D}^{2} - a^{*2})^{2} + \frac{2\lambda^{2}}{Pr}(\zeta\overline{D}^{3} - a^{*2}\zeta\overline{D} + 2a^{*2})]w^{*} = a^{*2}(\theta^{*} - c^{*}). \end{split}$$

The Darcy-Rayleigh number $R_{\rm m}^* \left(= {\rm g} \beta_{\rm m} \Delta C \Pi H / (\kappa \nu)\right)$ is based on the mushy-layer thickness H, where $\beta_{\rm m} = \beta - \alpha \Gamma$, $\Delta T = \Gamma \Delta C = T_{\rm L}(C_{\infty}) - T_{\rm E}$, $\Delta C = C_{\infty} - C_{\rm E}$, and Γ is the slope of the liquidus curve. The thermal Rayleigh number $R_{\rm T}^*$ and the solutal Rayleigh number $R_{\rm S}^*$ are defined as $R_{\rm T}^* = {\rm g} \Delta \Delta T H^3 / (\kappa \nu)$ and $R_{\rm S}^* = {\rm g} \beta \Delta C H^3 / (\kappa \nu)$, respectively. The parameter St is the Stefan number $(=\overline{L}/(C_{\rm P}\Delta T))$, and γ is the concentration ratio $(=(C_{\rm S}-C_{\infty})/\Delta C)$, where \overline{L} denotes the latent heat of fusion, $C_{\rm D}$ the

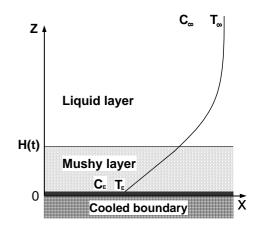


Fig. 1. Schematic diagram of liquid and mushy layers during time-dependent solidification.

specific heat, and C_s the solute concentration in solid. The parameter Le is the Lewis number $(=D/\kappa)$, and Pr is the Prandtl number $(=v/\kappa)$, where D is the solute diffusivity, and v is the kinematic viscosity. In the mushy layer, the basic-state equation is given by

$$\overline{D}^{2}\theta_{m0} + 2\lambda^{2}\zeta \left(1 + St \frac{\gamma - \theta_{i}}{(\gamma - \theta_{m0})^{2}}\right) \overline{D}\theta_{m0} = 0,$$

with $\theta_0=\theta_{\rm m0}$, $\overline{\rm D}\theta_0=\overline{\rm D}\theta_{\rm m0}$ at $\zeta=1$, and $\theta_{\rm m0}=-1$ at $\zeta=0$. In the liquid layer, the basic-state temperature and concentration fields are given by $\theta_0=\theta_{\rm s}+(\theta_{\rm i}-\theta_{\rm s}){\rm erfc}(\lambda\zeta)/{\rm erfc}(\lambda)$ and $c_0=\theta_{\rm i}{\rm erfc}(\lambda\zeta/\sqrt{Le})/{\rm erfc}(\lambda/\sqrt{Le})$, respectively, where $\theta_{\rm i}=(T_{\rm i}-T_{\rm i}(C_{\rm s}))/\Delta T$) is the temperature at the liquid-mush interface, and $\theta_{\rm s}=(T_{\rm i}-T_{\rm i}(C_{\rm s}))/\Delta T$) is the superheat. The following boundary conditions are applied to the self-similar stability equations:

for $\zeta \to \infty$, $\theta^* = 0$, $c^* = 0$, $w^* = 0$, $\overline{D}w^* = 0$,

at $\zeta=1$,

$$\begin{split} \boldsymbol{\theta}^* &= \frac{R_{\mathrm{T}}^*}{R_{\mathrm{S}}^*} \boldsymbol{c}^*, \quad \overline{\mathrm{D}} \boldsymbol{\theta}^* = R_{\mathrm{T}}^* \Bigg[\frac{\overline{\mathrm{D}} \boldsymbol{c}^*}{R_{\mathrm{S}}^*} - 2 \lambda^2 \bigg(\frac{1 - Le}{Le} \bigg) \overline{\mathrm{D}} \boldsymbol{\theta}_0 \boldsymbol{h}^* \Bigg], \quad \boldsymbol{\theta}^* = \frac{\boldsymbol{\Pi}^* R_{\mathrm{T}}^*}{R_{\mathrm{m}}^*} \boldsymbol{\theta}_{\mathrm{m}}^*, \quad \overline{\mathrm{D}} \boldsymbol{\theta}^* = R_{\mathrm{T}}^* \Bigg[\frac{\boldsymbol{\Pi}^*}{R_{\mathrm{m}}^*} \overline{\mathrm{D}} \boldsymbol{\theta}_{\mathrm{m}}^* + 2 \lambda^2 \bigg(\frac{St}{\boldsymbol{\theta}_{\mathrm{i}} - \gamma} \bigg) \overline{\mathrm{D}} \boldsymbol{\theta}_0 \boldsymbol{h}^* \Bigg], \\ \boldsymbol{w}^* &= \boldsymbol{w}_{\mathrm{m}}^* \boldsymbol{\Pi}^*, \quad \overline{\mathrm{D}} \boldsymbol{w}^* = 0, \quad \overline{\mathrm{D}} \boldsymbol{w}_{\mathrm{m}}^* = - \Bigg[\overline{\mathrm{D}}^3 \boldsymbol{w}^* - \boldsymbol{a}^{*2} \overline{\mathrm{D}} \boldsymbol{w}^* - \frac{2 \lambda^2}{Pr} (\overline{\mathrm{D}} \boldsymbol{w}^* - \overline{\mathrm{D}}^2 \boldsymbol{w}^*) \Bigg], \quad \boldsymbol{\chi}^* = \frac{\overline{\mathrm{D}} \boldsymbol{\theta}_{\mathrm{m0}}^*}{\boldsymbol{\theta}_{\mathrm{i}} - \gamma} \boldsymbol{h}^* \frac{R_{\mathrm{m}}^*}{\boldsymbol{\Pi}^*}, \end{split}$$

at $\zeta = 0$, $\theta_{\rm m}^* = 0$, $w_{\rm m}^* = 0$,

where Π^* (= Π/H^2) denotes the Darcy number. The Rayleigh numbers have the relations of $R_T^* = AR_m^*/\Pi^*$ and $R_S^* = (1+A)R_m^*/\Pi^*$, where $A = (-\Gamma\alpha/\beta_m)$ is the buoyancy ratio.

RESULTS AND DISCUSSION

The shooting method is employed to solve the self-similar stability equations [3]. We investigate the stability criteria for $A \rightarrow 0$, St=5, γ =20, Π^* =10⁻⁵, and Pr=10. These values are relevant to solidification of aqueous ammonium chloride solution. The marginal stability curves for various Lewis numbers with θ_{∞} =0.5 are shown in Fig. 2. The critical Darcy-Rayleigh number $R_{\rm m,c}^*$ for mushy-layer-mode convection is found to be 19 for θ_{∞} = 0.1 and Le = 0.013, and 7.6 for θ_{∞} = 1 and Le = 0.007. These $R_{\rm m,c}^*$ -values compare with the existing results [1,2,4]. The critical mushy-layer thickness H_c is predicted to be 0.8~2 cm for the onset of mushy-layer-mode convection, which is consistent with the existing experimental data [5].

The present results show that the Prandtl number, the Lewis number, and the superheat are destabilizing parameters for convective instabilities. With increasing the Darcy number Π^* , the critical Rayleigh numbers increase and mushy-layer mode convection becomes dominant. However, when Π^* is small, boundary-layer-mode convection becomes dominant, since the permeability of the mushy layer is small and the resistance to mushy-layer-mode convection is large. The critical time t_c to mark the onset of mushy-layer-mode convection is predicted from the relation of $t_c = H_c^2 / 4\lambda^2 \kappa$. It is found that

 $t_{\rm c}$ has a minimum point with varying θ_{∞} and that $t_{\rm c}$ decreases with increasing θ_{∞} for $\theta_{\infty} < 0.3$, while it increases for $\theta_{\infty} > 0.3$.

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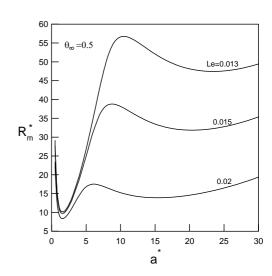


Fig. 2. Marginal stability curves for various Lewis numbers *Le* with $\theta_{\infty} = 0.5$.