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Onset of convection in a porous mush during binary solidification

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Abstract−The onset of convection in a mushy layer during solidification of a binary melt is investigated by using the propagation theory we have developed. The critical conditions for the mushy-layer-mode of instability are obtained numerically for aqueous ammonium chloride solution. The mush thickness at the onset of convection is predicted as a function of the solution viscosity and compared with the experimental data. The onset time of the mushy-layer-mode convection has a minimum point with varying the superheat. When the superheat is large, the mushy layer grows slowly and a long time is required for the onset of convection.

Key words: Onset of Convection, Solidification, Mush, Propagation Theory

INTRODUCTION

A mushy layer, made of dendritic crystals, is formed due to the morphological instability of the solid-liquid interface when a two-component melt is solidified. The mush is a region of the solid-liquid mixed phase and it is treated as a reactive porous medium. In the gravitational field, compositional convection can be induced by an unstable density profile in the mushy layer. Natural convection in the mushy layer causes freckles in solidified alloys, and hence the criteria for the convection are important in the crystal growth process for the semi-conductor industry [1,2].

Compositional convection in the mushy layer during solidification has attracted attention for several decades [3-13]. Experiments on natural convection in the mushy layer were performed by using aqueous ammonium chloride (NH_4Cl) solution, which is similar to metal alloys in solidification process. Chen and Chen [5] determined the critical conditions for the onset of plum convection in the mushy layer. Tait and Jaupart [6] found that chimneys are formed by natural convection in the mush. They estimated the critical conditions for the onset of plum convection in the mush with varying the viscosity of a melt. In experiments, small scale convection (the boundary-layer-mode of convection) first appears in the liquid layer, but it does not influence the fluid in the mush [6]. Theoretically, when the Lewis number Le ($=D/\kappa$) is small, where D is the solute diffusivity and κ is the thermal diffusivity, the mushy-layer-mode of convection predominates and the boundary-layer-mode is stabilized [7,10]. For Le$\rightarrow$0 Worster [7] investigated the mushy-layer-mode of instability and compared his results with Tait and Jaupart's [6] experimental results. Emms and Fowler [9] determined the critical mush Rayleigh number by using the quasi-static stability analysis.

In this work, the onset of convection in the mushy layer during time-dependent solidification of a binary melt cooled from below is investigated by using the propagation theory we have developed [14-20]. In the previous work [15] the Lewis number was considered to investigate the boundary-layer-mode of instability. Here, we treat the asymptotic form of the stability equations in the limit of Le$\rightarrow$0 in order to study the mushy-layer-mode of instability. Using the self-similar stability analysis, the critical mush thickness and the onset time of convection in the mushy layer are predicted for solidification of aqueous ammonium chloride solution.

SELF-SIMILAR STABILITY EQUATIONS

When a two-component melt is solidified, thermal and compositional fields develop in the liquid and mushy layers. The governing equations for the present study are the same as those in the previous paper [15], and here the details of the linear stability analysis are not written. A schematic diagram of the solidifying mush is shown in Fig. 1. A binary melt is initially quiescent at a temperature T_∞ and a solute (NH_4Cl) concentration C_∞. At the time t=0 the melt is suddenly cooled from the bottom boundary. Then the mushy layer grows from below, and compositional convection may be induced by the light residual liquid (e.g., water in aqueous ammonium chloride solution). The mush thickness H ($=2\lambda\sqrt{\kappa t}$) is a function of time t, where λ is the phase-change rate and κ is the thermal diffusivity. The bottom boundary of the mush is at the eutectic temperature T_E and the eutectic composition C_E. After linear stability theory is ap-

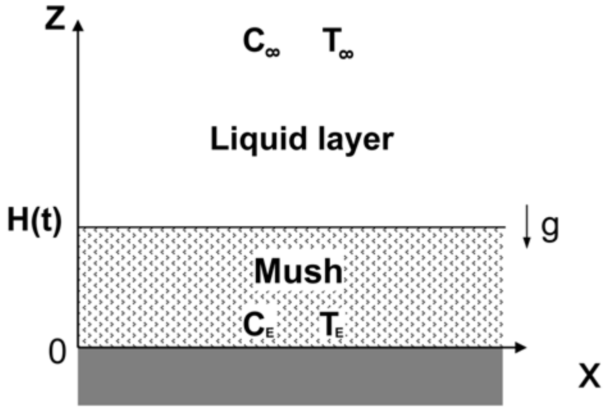

Fig. 1. Schematic diagram of mush solidifying from below.

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plied to the two sets of the dimensionless governing equations for the liquid and mushy layers, the perturbation equations are transformed by using self-similarity.

In the propagation theory the time-dependent disturbance equations are transformed to functions of a similarity variable ζ (=z/h), where z is the vertical distance, h ($=2\lambda\tau^{1/2}$) the mushy-layer thickness, τ the time scaled by L^2/κ, and L an arbitrary length. The Lewis number Le represents the relative magnitude of the compositional boundary-layer thickness to the thermal boundary-layer thickness. In order to study the mushy-layer-mode of convection we use the asymptotic form of the self-similar stability equations for the limiting case of Le=0. In this case, the concentration field in the liquid layer is ignored and the self-similar stability equations are obtained as follows:

$$(\overline{D}^2+2\lambda^2\zeta\overline{D}-a^{*2})\theta^*=\frac{AR_m^*}{\Pi^*}w^*\overline{D}\theta_0, \tag{1}$$

$$\left[(\overline{D}^2-a^{*2})^2+\frac{2\lambda^2}{Pr}(\zeta\overline{D}^3-a^{*2}\zeta\overline{D}+2a^{*2})\right]w^*=a^{*2}\theta^*. \tag{2}$$

In the mushy layer the self-similar stability equations are

$$(\overline{D}^2+2\lambda^2\zeta\overline{D}-a^{*2})\theta_m^*=R_m^*w_m^*\overline{D}\theta_{m0}-2\lambda^2 St\zeta\overline{D}\chi^*, \tag{3}$$

$$2\lambda^2\zeta[\chi^*\overline{D}\theta_{m0}+\chi_0\overline{D}\theta_m^*+(\theta_{m0}-\gamma)\overline{D}\chi^*+\theta_m^*\overline{D}\chi_0]=R_m^*w_m^*\overline{D}\theta_{m0}, \tag{4}$$

$$(\overline{D}^2-a^{*2})w_m^*=a^{*2}\theta_m^*, \tag{5}$$

where $\overline{D}$ represents $d/d\zeta$ and a^* is the horizontal wave number. The vertical velocity disturbance w^* is scaled by $\kappa H^2/L^3$ and w_m^* by $\kappa\Pi/L^3$, where Π is the permeability of the mush. The parameter Pr is the Prandtl number ($=\nu/\kappa$), St the Stefan number ($=\overline{L}/(C_P\Delta T)$), and γ the concentration ratio ($=(C_s-C_\infty)/\Delta C$)), where ν is the kinematic viscosity, $\overline{L}$ the latent heat of fusion, C_p the specific heat, C_s the solute concentration in solid, $\Delta T=\Gamma\Delta C=T_L(C_\infty)-T_E$, and Γ the slope of the liquidus curve. The Darcy-Rayleigh number R_m^* is defined as

$$R_m^*=\frac{g\beta_m\Delta C\Pi H}{\kappa\nu}, \tag{6}$$

where g denotes the gravity acceleration, $\beta_m=\beta-\alpha\Gamma$, and α and β are the thermal and solutal expansion coefficients, respectively. The Darcy number Π^* is defined as Π/H^2 and A ($=\Gamma\alpha/\beta_m$) is the buoyancy ratio. The temperature disturbance θ^* is scaled by $\kappa\nu/(g\alpha L^3)$, θ_m^* by $\kappa\nu\Gamma/(g\beta_m L^3)$, and the porosity disturbance χ^* by $\kappa\nu/(g\beta_m\Delta CL^3)$.

The basic-state equations and boundary conditions are given by

$$\frac{d^2\theta_0}{d\zeta^2}+2\lambda^2\zeta\frac{d\theta_0}{d\zeta}=0 \qquad \text{for } \zeta>1, \tag{7}$$

$$\frac{d^2\theta_{m0}}{d\zeta^2}+\left[1+\frac{St}{\gamma}\left(\frac{1}{1-\frac{\theta_{m0}}{\gamma}}\right)^2\right]2\lambda^2\zeta\frac{d\theta_{m0}}{d\zeta}=0 \qquad \text{for } \zeta<1, \tag{8}$$

$$\theta_0=\theta_\infty \qquad \text{for } \zeta\to\infty, \tag{9}$$

$$\theta_0=\theta_{m0}=0,\ \frac{d\theta_0}{d\zeta}=\frac{d\theta_{m0}}{d\zeta} \qquad \text{at } \zeta=1, \tag{10a,b}$$

$$\theta_{m0}=-1 \qquad \text{at } \zeta=0, \tag{11}$$

where the superheat θ_∞ is defined as $(T_\infty-T_L(C_\infty))/\Delta T$. The porosity profile in the mushy layer is given by $\chi_0=\gamma/(\gamma-\theta_{m0})$.

The linearized boundary conditions are applied to the self-similar stability equations:

for $\zeta\to\infty$

$$\theta^*=w^*=\overline{D}w^*=0, \tag{12}$$

at $\zeta=1$

$$\theta^*=A\theta_m^*,\ \overline{D}\theta^*=A\overline{D}\theta_m^*, \tag{13a,b}$$

$$w^*=w_m^*\Pi^*,\ \overline{D}w^*=0,\ \chi^*=0, \tag{13c-e}$$

$$\overline{D}w_m^*=-\left[\overline{D}^3w^*-a^{*2}\overline{D}w^*-\frac{2\lambda^2}{Pr}(\overline{D}w^*-\overline{D}^2w^*)\right], \tag{14}$$

at $\zeta=0$

$$\theta_m^*=w_m^*=0. \tag{15}$$

RESULTS AND DISCUSSION

We set the dimensionless parameters St=5, γ=20, $\Pi^*=10^{-5}$, and Pr=10, which are appropriate for aqueous ammonium chloride solution. When the buoyancy ratio A is very small (we set $A=10^{-10}$), the effect of the thermal buoyancy force is neglected. For a given θ_∞, the minimum R_m^*-value ($R_{m,c}^*$) and its corresponding wave number a_c^* are found numerically. The shooting method is employed to solve the self-similar stability equations [15]. For $\Pi^*<10^{-4}$, $R_{m,c}^*$-value is not influenced by the Darcy number Π^*.

Fig. 2 shows the marginal stability curve for θ_∞=0.5. The critical values ($R_{m,c}^*$=10.75 and a_c^*=1.57) determine the conditions to mark the onset of convection in the mushy layer. The parameter θ_∞ represents the initial temperature of the melt or the superheat, which is the ratio of temperature differences in the liquid and mushy layers. In the existing experiment [6] the value of θ_∞ ranges from 0.1 to 1.1, and here θ_∞=0.5 is representative of the experiment. The streamlines of a two-dimensional convective roll-cell at the critical condition for θ_∞=0.5 are plotted in Fig. 3. It is seen that the maximum

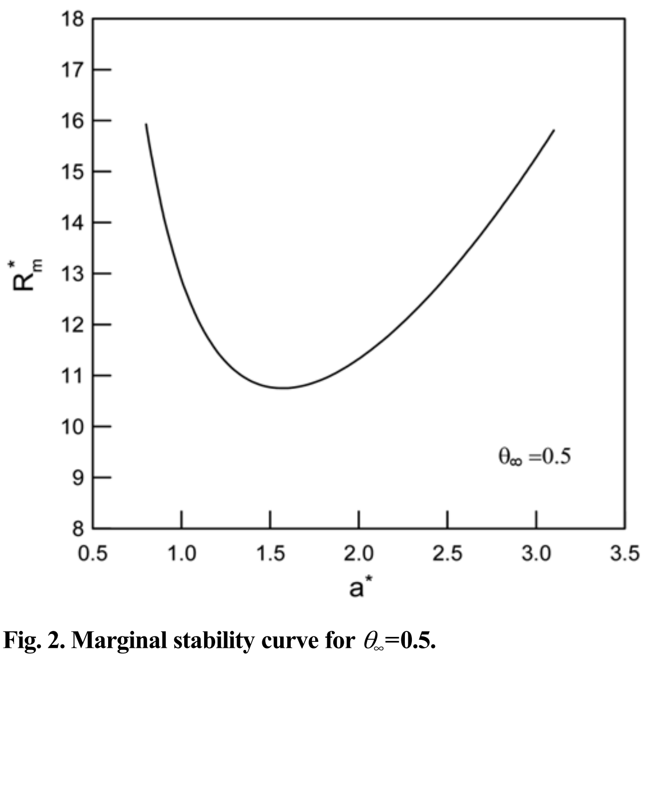

Fig. 2. Marginal stability curve for θ_∞=0.5.

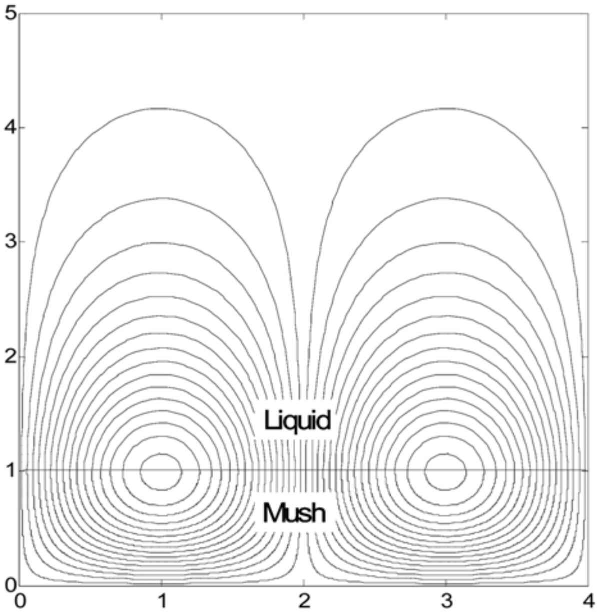

Fig. 3. Streamlines of two-dimensional convective roll for θ_∞=0.5.

velocity is at the liquid-mush interface and the critical flow appears mainly within the distance of 5 times the mush thickness. The convective upflow between two cells in the mush causes a local decrease of the solid fraction, resulting in the formation of chimneys. The upflow through the chimney develops to the plum convection, a strong upwelling from the mush.

The critical Rayleigh numbers for the mushy layer obtained from the linear stability theory were compared with the experimental results in the previous papers [7,9,14]. In the present study $R^*_{m,c}$-values are found for different values of the superheat θ_∞, and the critical mush thickness H_c at the onset of mushy-layer-mode of convection is predicted. Tait and Jaupart [6] observed the plum convection at the same time when porosity fluctuations occur in the mush and measured the mush thicknesses at this moment. For the lowest superheats they determined a value of 25 for the critical Darcy-Rayleigh number using the data of the mush thickness with different viscosities of NH_4Cl solution in Fig. 4 of their paper. Based on the experimental value of R^*_m=25 we calculate the critical mush thickness H_c at the onset of the mushy-layer-mode of convection from the relation of $H_c \propto \mu$ in Eq. (6), where μ is the solution viscosity. The present $R^*_{m,c}$-values are 8.7-19 for $1.1 \geq \theta_\infty \geq 0.1$, and H_c is plotted as a function of the solution viscosity in Fig. 4. The experimental results are higher than the present H_c-values. The propagation theory predicts the critical condition for the onset of convection at which disturbances begin to grow. Therefore a time delay exists between the convective instability and the onset of channeling in the mush. The critical conditions from experimental observation of the onset of convection are expected to be higher than the theoretical prediction [9].

For St=5 and γ=20, the values of λ obtained numerically from Eqs. (7)-(11) are shown in Fig. 5. The present values of λ are very close to Emms and Fowler's [9] in Fig. 3 of their paper. For $\gamma \gg \theta_{m0}$ the basic-state Eq. (8) becomes $d^2\theta_{m0}/d\zeta^2+(1+St/\gamma)2\lambda^2\zeta d\theta_{m0}/d\zeta=0$, which is similar to Emms and Fowler's [9] model for ammonium chloride solution. The critical time τ_c to mark the onset of mushy-layer-mode of convection is predicted from the relation of $\tau_c R_m^2 = R^{*2}_{m,c}/(2\lambda)^2$, where $R_m = g\beta_m \Delta T \Pi L/(\Gamma \kappa \nu)$. From Fig. 5 and the present $R^*_{m,c}$-values, $\tau_c R_m^2$ is plotted as a function of θ_∞ in Fig. 6. The critical Darcy-Rayleigh number is found to decrease as θ_∞ increases, and it is seen that the onset time of convection τ_c follows this trend for $\theta_\infty \leq 0.27$. It is of interest that τ_c has a minimum point with varying θ_∞, and increasing θ_∞ appears to delay τ_c for $\theta_\infty \geq 0.27$ or $\lambda \leq 0.82$. However, the change of τ_c is relatively not much for the range of $0.1 \leq \theta_\infty \leq 1$. When θ_∞ is large, the phase-change rate λ is small and the mushy layer grows slowly so that a large time is required for the onset of convection. For $\lambda \rightarrow 0$, the basic temperature profile becomes linear in the mushy layer and Eqs. (1)-(5) are similar to the instability problem of the superposed liquid and porous layers

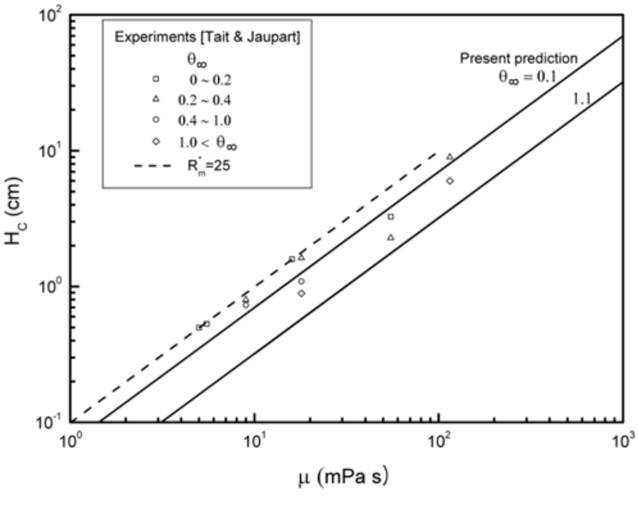

Fig. 4. Critical mush thickness H_c as a function of viscosity of aqueous ammonium chloride solution. Tait and Jaupart [6] determined R^*_m=25 (the dashed line) for the lowest superheats. The present $R^*_{m,c}$-value is 19 for θ_∞=0.1 and 8.7 for θ_∞=1.1, shown by the solid line.

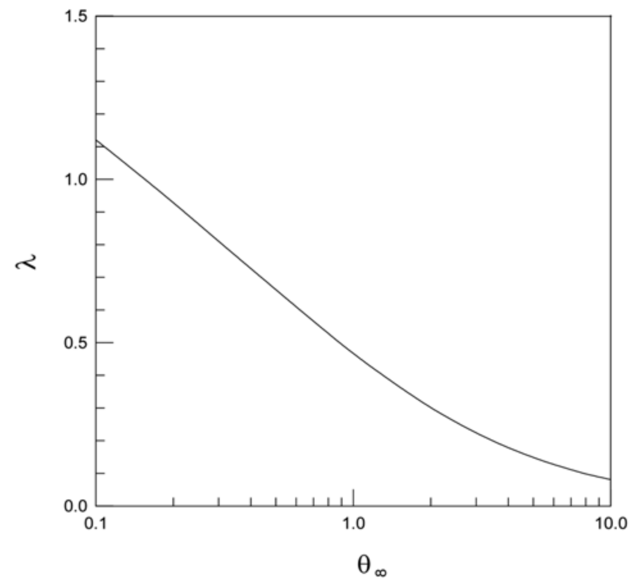

Fig. 5. Variation of λ with superheat θ_∞ for St=5 and γ=20.

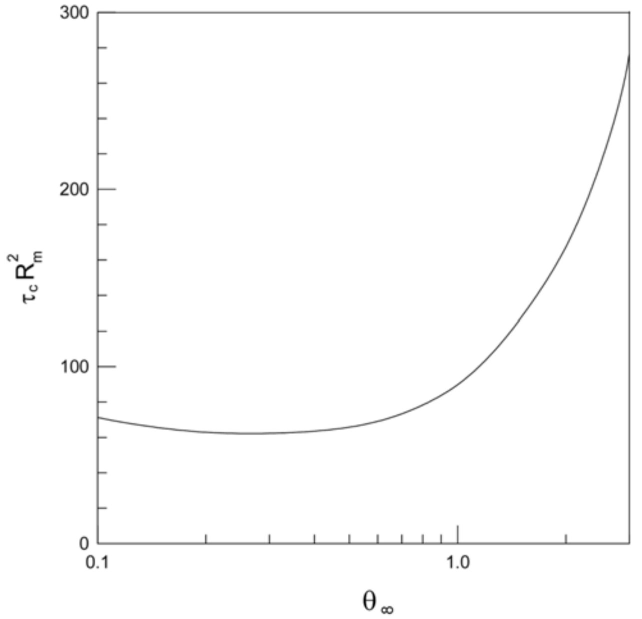

Fig. 6. Variation of $\tau_c R_m^2$ with superheat θ_∞.

with a linear temperature profile.

CONCLUSION

The critical conditions to mark the onset of convection have been found numerically for solidification of a mushy layer cooled from below. Based on the propagation theory, the self-similar stability equations for the liquid and mushy layers are employed when the concentration field in the liquid layer is not considered. The critical Darcy-Rayleigh numbers for the mushy-layer-mode of convection are found for different values of the superheat θ_∞. The critical mush thicknesses are predicted to be lower than the existing experimental results for solidification of aqueous ammonium chloride solution. The onset time of convection has a minimum point with varying θ_∞ and increases with θ_∞ for $\theta_\infty \geq 0.27$.

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