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Technical Note

Effects of solid layer thickness and nominal composition on double-diffusive instabilities during solidification of a binary alloy cooled from the top

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1. Introduction

When a horizontal layer of fluid is cooled and solidified from the top, a cellular form of natural convection may occur inside the liquid, similar to that observed during classical Rayleigh–Benard convection. As a result of the interaction between the cellular convection and directional solidification, the phase-change interface tends to get wavy. The above situation can be mathematically represented by considering two horizontal parallel plates at $z = 0$ and $z = h$. The lower plate at $z = h$ is fixed at temperature $T = T_1$ and the upper plate at $z = 0$ is kept at a temperature $T = T_0$. Due to occurrence of solidification on account of cooling from the top, there is a solid–liquid interface at $z = \eta (0 \leqslant \eta \leqslant h)$, which is assumed to be at a quasi-steady state. The physical situation is shown schematically in Fig. 1. As a result of interface perturbation on account of convection occurring below, the interface is likely to assume an irregular shape. It can be noted here that when the solidifying medium is a binary alloy that solidifies non-isothermally, the natural convection in the liquid is double-diffusive in nature (as a consequence of concentration and temperature gradients prevailing in the solidifying domain). Instabilities in thermo-solutal convection in such cases have attracted considerable attention in the literature [1,2]. However, the effects of nominal alloy composition and solid layer thickness on associated double-diffusive instabilities are yet to be addressed in the literature, to the best of our knowledge.

The aim of the present study is to investigate the effects of solid layer thickness and nominal composition on the double-diffusive instabilities, as a binary alloy of any specified initial composition is directionally solidified from the top. However, it can be noted here that in typical alloy solidification problems, there is also a susceptibility of the advancing solid–liquid interface to morphological instabilities in which the initially planar phase boundary becomes deformed, leading to a cellular pattern of microsegregation and ultimately a dendritic growth structure. The segregation, in turn, can generate gradients of concentration, which may themselves cause convective motions, thereby changing the growth conditions at the interface. However, in the regimes of practical interest, the coupling between the morphological and convective instability mechanisms may be rather weak [3] due to the widely differing spatial scales involved. In this paper, we specifically aim to investigate the effects of bulk-liquid double-diffusive convection on convective instabilities during the solidification process, as a binary alloy is cooled and solidified from the top. In order to facilitate an analytical treatment, the mushy zone (a diffused region constituting of solid dendrites and interdendritic liquid) thickness is assumed to be negligible in comparison to the solid layer thickness. The above assumption decouples the mushy zone flow from the bulk flow, but incorporates the effects of a continuously deforming solidification front under a quasi-steady situation. Another simplifying assumption is made in that the property variation within each phase is neglected. The double-diffusive convective instability theory is coupled with interface perturbations that enables us to obtain critical Rayleigh numbers of flow transition during thermo-solutal convection in the presence of a deformable solidification interface. It is important to mention that in the present study, only direct modes of

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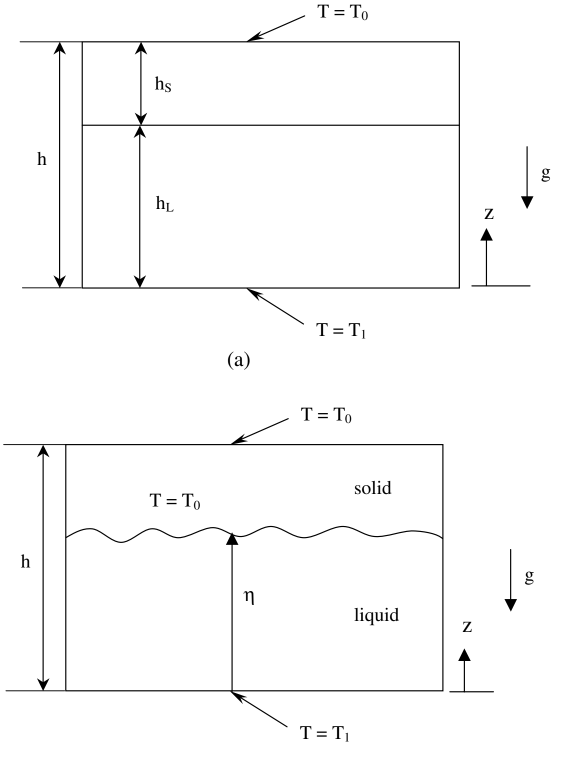

(a)

(b)

Fig. 1. A schematic diagram of the model system under investigation for double diffusive convection in the liquid: (a) unperturbed state and (b) perturbed state.

instabilities are considered, since the other significant mode of instability, characterised by overstable oscillations, is discussed in details elsewhere [4]. However, such overstable oscillations are the first to occur in a double-diffusive system only in a specific situation when the salinity field is statically stable [4].

2. Analysis and results

Mathematical modeling is performed for the physical situation represented in Fig. 1. The governing equation for the solid region of interest is the heat conduction equation. In this case, we neglect species diffusion in the solid, since typical solid-state mass diffusivities are much lower than the corresponding thermal diffusivities. Also, species mass diffusion coefficient in liquid is about three orders of magnitude higher than that in the solid. Energy conservation equation for the solid can be stated as

$$\frac{\partial T_S^*}{\partial t^*} = \alpha \nabla^2 T_S^* \tag{1}$$

where α_S represents thermal diffusivity of solid phase, and t is the variable time. However, for analysis of transport phenomena in the liquid region, the mass, momentum, energy, as well as species conservation equations have to be invoked as

$$\nabla \cdot \vec{u} = 0 \tag{2}$$

$$\frac{\partial u}{\partial t} + \vec{u} \cdot \nabla u = -\frac{1}{\rho_L}\frac{\partial p}{\partial x} + \upsilon \nabla^2 u \tag{3a}$$

$$\frac{\partial v}{\partial t} + \vec{u} \cdot \nabla v = -\frac{1}{\rho_L}\frac{\partial p}{\partial y} + \upsilon \nabla^2 v \tag{3b}$$

$$\begin{aligned}\frac{\partial w}{\partial t} + \vec{u} \cdot \nabla w = -\frac{1}{\rho_L}\frac{\partial p}{\partial z} + \upsilon \nabla^2 w + g\{\beta_T(T - T_{\mathrm{ref}}) \\ + \beta_S(C_l - C_{\mathrm{ref}})\}\end{aligned} \tag{3c}$$

$$\frac{\partial T_L}{\partial t} + \vec{u} \cdot \nabla T_L = \alpha_L \nabla^2 T_L \tag{4}$$

$$\frac{\partial C_L}{\partial t} + \vec{u} \cdot \nabla C_L = D_l \nabla^2 C_L \tag{5}$$

In the above equations, u, v and w represent components of the velocity vector $\vec{u}$ along x, y, z, respectively. The symbol T represents temperature, C represents solute concentration, p is the pressure, β_S is the volumetric coefficient of solutal buoyancy, β_T represents volumetric coefficient of thermal buoyancy, α_L represents thermal diffusivity of the liquid phase, and represents kinematic viscosity. In the mathematical analysis presented in this paper, the subscripts 'L' and 'S' refer to liquid and solid phases, respectively.

For convenience in the analytical treatment, we may suitably non-dimensionalise the above governing equations coupled with appropriate boundary conditions. The non-dimensional quantities are denoted by a superscript '*'. The reference length scale is chosen as h_L (height of the liquid layer), time scale is chosen as h_L^2/α_L, velocity scale is chosen as $\widehat{w}$ and pressure scale is chosen as $\upsilon_l \rho_l \widehat{w}_l / h_L$, where $\widehat{w}$ is given by

$$\widehat{w} = \{g\beta_T(T_0 - T_{\mathrm{ref}})h_L \alpha_L/\gamma_L\}^{0.5} \tag{6}$$

Further, the solid and liquid state temperatures are non-dimensionalised as

$$T_L^* = (T_L - T_{\mathrm{sol}})/(T_0 - T_{\mathrm{sol}}) \tag{7}$$

$$T_S^* = (T_S - T_{\mathrm{sol}})/(T_0 - T_{\mathrm{sol}}) \tag{8}$$

where T_{sol} is the solidus temperature. In the above equations, the subscripts '0' and 'sol' refer to the initial condition and the solidus, respectively.

Similarly, the liquid state composition is non-dimensionalised as

$$C_L^* = (C_L - C_{\mathrm{sol}})/(C_0 - C_{\mathrm{sol}}) \tag{9}$$

With the parameters defined as above, the non-dimensionalised governing equations become

Energy conservation equation for solid phase

$$\frac{\partial T_S^*}{\partial t^*} = \alpha \nabla^2 T_S^* \tag{10}$$

where $\alpha = \alpha_S/\alpha_L$.

Continuity equation for the liquid phase

$$\nabla \cdot \vec{u}^* = 0 \tag{11}$$

Momentum conservation equations in the liquid phase

$$Pr^{-1}\left(\frac{\partial u^*}{\partial t^*} + R_T^{0.5}\vec{u}^* \cdot \nabla u^*\right) = -\frac{\partial p^*}{\partial x^*} + \nabla^2 u^* \tag{12a}$$

$$Pr^{-1}\left(\frac{\partial v^*}{\partial t^*} + R_T^{0.5}\vec{u}^* \cdot \nabla v^*\right) = -\frac{\partial p^*}{\partial y^*} + \nabla^2 v^* \tag{12b}$$

$$Pr^{-1}\left(\frac{\partial w^*}{\partial t^*} + R_T^{0.5}\vec{u}^* \cdot \nabla w^*\right) = -\frac{\partial p^*}{\partial z^*} + \nabla^2 w^* + R_T^{0.5} T_L^* + N R_T^{0.5} C_L^* \tag{12c}$$

where $Pr = \upsilon_L/\alpha_L$ and $N = \{\beta_S(C_0 - C_{sol})\}/\{\beta_T(T_0 - T_{sol})\}$ = buoyancy ratio.

Energy conservation equation in the liquid phase

$$\frac{\partial T_L^*}{\partial t^*} + R_T^{0.5}\vec{u}^* \cdot \nabla T_L^* = \nabla^2 T_L^* \tag{13}$$

where $R_T = g\beta_T(T - T_{sol})h_1^3/\upsilon_L\alpha_L$.

Species conservation equation in liquid phase

$$\frac{\partial C_L^*}{\partial t^*} + R_T^{0.5}\vec{u}^* \cdot \nabla C_L^* = \frac{D_L}{\alpha_L}\nabla^2 C_L^* \tag{14}$$

The non-dimensional boundary conditions can be stated as

$$u^* = v^* = w^* = 0, \quad T_L^* = 1, \quad C_L^* = 1 \quad \text{at } z^* = 0 \tag{15}$$

$$u^* = v^* = w^* = 0, \quad T_s^* = (T_1 - T_{sol})/(T_0 - T_{sol}), \quad \text{at } z^* = 1 + r \tag{16}$$

where $r = h_S/h_L$.

Energy balance at the interface

$$\rho St \frac{\partial \eta^*}{\partial t^*} = \{k\nabla T_S^* - \nabla T_L^*\} \cdot \hat{n} \quad \text{at } z^* = \eta^* \tag{17}$$

where $\rho = \rho_S/\rho_L$, $k = k_S/k_L$ and $St = \{\rho_L L \alpha_L\}/\{k_L(T_0 - T_{sol})\}$, L being the latent heat of fusion.

Solute balance at the interface

$$[1 - k_p][C_L^*(C_0 - C_{sol}) + C_{sol}]\frac{\partial \eta^*}{\partial t^*} = -(C_0 - C_{sol})\nabla C_L^* \cdot \hat{n} \quad \text{at } z^* = \eta^* \tag{18}$$

where C_{sol} is the solid composition corresponding to the phase-change temperature, and k_p is the partition-coefficient.

$$T_L^* = T_S^* = 0 \quad \text{at } z^* = \eta^* \tag{19}$$

Kinetic condition for interface deformation

$$\frac{(1-\rho)\frac{\partial \eta^*}{\partial t^*}}{\{1 + (\partial\eta^*/\partial x^*)^2 + (\partial\eta^*/\partial y^*)^2\}^{0.5}} = \vec{u}^* \cdot \hat{n} \quad \text{at } z^* = \eta^* \tag{20}$$

No-slip conditions at the interface

$$\vec{u}^* \cdot \hat{t}_1 = 0 \quad \text{at } z^* = \eta^* \tag{21a}$$

$$\vec{u}^* \cdot \hat{t}_2 = 0 \quad \text{at } z^* = \eta^* \tag{21b}$$

where $\hat{t}_1$ and $\hat{t}_2$ are unit vectors in two tangential directions with respect to the interface, mutually orthogonal to each other.

For the subsequent perturbation analysis, the superscript '*' is dropped from the dimensionless quantities for convenience, but the dimensionless quantities are used only.

First, the base state solutions are obtained, denoted by the subscript 'B'. At the basic state, all velocity components are zero and the pressure distribution within the fluid is hydrostatic. The basic state temperature profile within the solid is first solved to yield

$$T_{S,B} = k^{-1}(1 - z) \tag{22}$$

Similarly, the liquid phase temperature profile at the basic state is obtained as

$$T_{L,B} = 1 - z \tag{23}$$

Proceeding in a similar way, we obtain

$$C_{L,B} = 1 - z \tag{24}$$

In the subsequent formulation, instabilities associated with quasi-steady double diffusive convection in the liquid phase are considered. In a physical sense, the aim is to consider a basic state in which a slight rise in the temperature of the upper region causes a slight melting of the solid, leading to a perturbation at the interface. Regarding the perturbed equations, a theory first described by Malkus and Voronis [5] can be followed as:

$$\phi = \phi_B + \varepsilon\phi_1 + \varepsilon^2\phi_2 \tag{25}$$

where ϕ is a generic form for either of $\vec{u}$, p, T_L, T_S, C_L, R_T, and ε is a small number.

In Eq. (25), all the variables are assumed to be of the form $\hat{f}(z)\phi(x,y)$, where $\hat{f}$ and ϕ are two separable generic functions of z and (x, y), respectively. One can expand the non-dimensional governing equations according to the above assumptions, and subsequently impose the boundary conditions. In essence, we aim to perform a normal-mode analysis. Regarding the interface boundary conditions, it is important to note that the associated variables are to be expanded in a Taylor's series about $z = 1$ (i.e., basic location of the interface). Accordingly, we obtain

$$\widehat{T}_{S1} = k^{-1}\widehat{\eta}_1 \frac{\sinh\{ar(1-\xi)\}}{\sinh(ar)} \tag{26}$$

where a is the wave number of perturbation and $\xi = (z-1)/r$.

The liquid-state temperature-field, however, cannot be solved without invoking the momentum and species conservation equations, since they are coupled together. The appropriate interface boundary conditions for temperature distribution in the liquid can be obtained by using the Stefan boundary condition (Eq. (17)) and temperature distribution inside the solid (given by Eq. (26)) to yield

$$\widehat{T}_{L1} = \widehat{\eta}_1 \quad \text{at } z = 1 \tag{27}$$

$$D\widehat{T}_{L1} = -a\widehat{\eta}_1 \cosh(ar) \quad \text{at } z = 1 \tag{28}$$

The linear stability of the problem in the liquid under neutral stability conditions is posed by obtaining a system of self-adjoint equations described below. The following equations are basically derived by equating O(ε) terms in the governing perturbation equations. Accordingly, one gets

$$\nabla^2 T_{L1} + R_{T0}^{0.5} w_1 = 0 \tag{29}$$

$$-\frac{\partial p_1}{\partial y} + \nabla^2 v_1 = 0 \tag{30}$$

$$-\frac{\partial p_1}{\partial x} + \nabla^2 u_1 = 0 \tag{31}$$

$$-\frac{\partial p_1}{\partial z} + \nabla^2 w_1 + R_{T0}^{0.5} T_{L1} + R_{T0}^{0.5} N C_{L1} = 0 \tag{32}$$

$$\frac{D_L}{\alpha_L} \nabla^2 C_{L1} + R_{T0}^{0.5} w_1 = 0 \tag{33}$$

$$\{(D^2 - a^2)^3 - R_{0,\mathrm{eq}} a^2\}\widehat{w}_1 = 0 \tag{34}$$

where

$$R_{0,\mathrm{eq}} = R_{T0} + N\frac{\alpha_L}{D_L} R_{T0} = R_{T0} + R_{S0} \tag{35}$$

In the above equation, R_{S0} is the solutal Rayleigh number.

The appropriate boundary conditions for Eq. (35) are

$$\widehat{w}_1 = D\widehat{w}_1 = (D^2 - a^2)\widehat{w}_1 = 0 \quad \text{at } z = 0 \tag{36a}$$

$$\widehat{w}_1 = D\widehat{w}_1 = 0, \quad D\{(D^2 - a^2)\widehat{w}_1\} + a\coth(ra)\{(D^2 - a^2)^2\widehat{w}_1\} = 0 \quad \text{at } z = 1 \tag{36b}$$

The solvability conditions for the above system gives rise to a critical Rayleigh number (R_{cr}) as a function of r, where R is the algebraic sum of thermal and solutal Rayleigh numbers, and r is defined as the ratio of height of the solid layer to the liquid layer. The critical Rayleigh number is plotted as a function of r and the critical wave number (a_{cr}) is also plotted as a function of r in Fig. 2(a) and (b), respectively. It can be seen from the figures that the critical Rayleigh number for onset of instability decreases asymptotically as the relative height

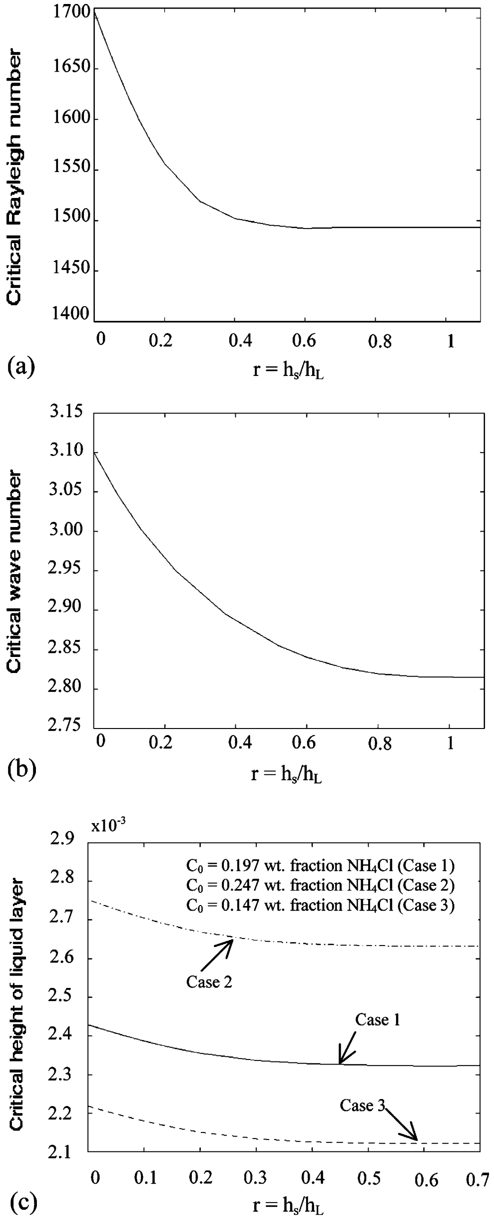

Fig. 2. Critical parameters for double diffusive convection in bulk liquid: (a) variation with critical Rayleigh number with respect to r; (b) variation with critical wave number with respect to r and (c) variation of critical liquid layer height (for onset of instability) with r, for the cases in which solutal buoyancy is absent (case 1), opposes thermal buoyancy (case 2), and aids thermal buoyancy (case 3).

of the solid layer increases. In other words, double diffusive instabilities are associated with lower values of minimum liquid-layer heights, in comparison to the case where no phase change can occur. This can be attributed to the fact that the interface perturbation acts as an additional source of disturbance in the present case, which can trigger off instabilities more easily. However, it is important to note that the critical Rayleigh number is actually a combination of the thermal and solutal Rayleigh numbers. Thus, the corresponding critical height not only depends on the relative depth of the solid layer, but also depends on the relative signs of β_T and β_S, which essentially decide whether thermal buoyancy effects are aided by solutal buoyancy effects, or vice-versa. Hence, for a particular value of r, the critical height of the liquid layer would be dependent on the initial concentration of the mixture. Such effects are illustrated for an NH_4Cl–H_2O system in Fig. 2(c), where the nominal compositions are taken as the eutectic composition (case 1), greater than eutectic concentration by 5 wt.% (case 2), and lesser than the eutectic concentration by 5 wt.% (case 3), respectively. It can be noted that case 2 leads to a situation where solutal buoyancy opposes the thermal buoyancy, whereas in case 3 they aid each other. Case 1 does not involve any effects of solutal buoyancy at all. It can be seen from Fig. 2(c) that the critical height of liquid layer for case 3 is the lowest, and the same is the highest for case 2. This can be attributed to the fact that since the thermal and solutal buoyancy effects aid each other in case 3, convection does not die out easily and continues to prevail for a much longer time, as compared to the other cases. During this time, the solidification front progresses considerably, leading to a reduced critical height of the liquid layer. On the other hand, in case 2, the solutal buoyancy opposes the thermal buoyancy, and hence the convection is expected to die down much earlier, resulting in a much increased critical depth of the liquid layer than the third case. Case 1 results in an isothermal solidification very much alike a single-component system, with no effects of solutal buoyancy. Accordingly, the critical height of the liquid layer falls in between the corresponding values for case 2 and case 3.

Appendix A. Analysis of planform functions and first-order variations at criticality

For an illustration, we consider the solidification of an eutectic binary system, as it is cooled from the top. After separation of the dependent variables (such as velocity, temperature), we obtain a governing equation for the planform function (ϕ) as

$$\frac{\partial^2\phi}{\partial x^2} + \frac{\partial^2\phi}{\partial y^2} = -a^2\phi \qquad \text{(A.1)}$$

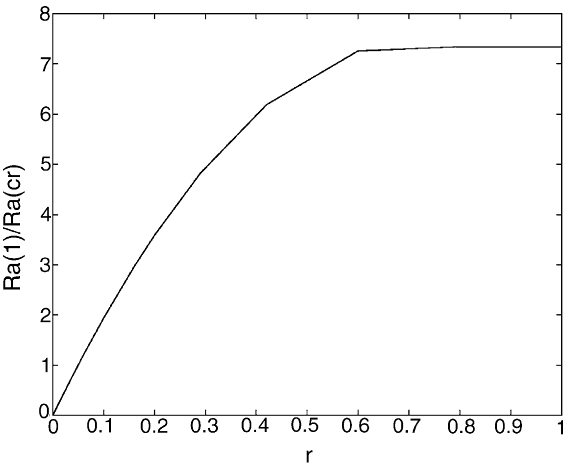

Fig. 3. Variation of first-order correction of Ra with 'r'.

under the normalization condition

$$\bar{\phi}^2 = 1 \qquad \text{(A.2)}$$

where the 'bar' over ϕ denotes the horizontal average over one period in x and y; i.e., over one cell. In Eq. (A.1), a is the wave number. Stability considerations [6] lead to the appearance of either roll-type or hexagonal cells. The occurrence of the type of cells is determined by the following special form of ϕ:

$$\phi(x,y) = Y\cos\frac{ky}{2}\cos\frac{\sqrt{3}kx}{2} + Z\cos ky \qquad \text{(A.3)}$$

In the above equation, rolls have $Y = 0$, $Z \neq 0$ and hexagons have $Y = \pm 2Z$. Under these conditions, the normalization constraint (A.2) requires

$$\frac{Y^2}{4} + \frac{Z^2}{2} = 1 \qquad \text{(A.4)}$$

so that $Z = \pm\sqrt{2}$ for the roll patterns and $Z = \pm\sqrt{\frac{2}{3}}$ for the hexagonal pattern.

In order to obtain a deeper insight into the effects of the resultant eigenfunctions on the first-order correction of Ra (say, $Ra(1)$), the ratio $Ra(1)/Ra_{cr}$ can be plotted as a function of $r(= h_S/h_L)$, as depicted in Fig. 3, with a normalization of $\max \widehat{w}_1(z) = 0.94a_c^2(r)$, where a_c is the critical wave number.

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