Supplementary material

Role of microstructure and composition on the natural convection during ternary alloy solidification

Virkeshwar Kumar, Atul Srivastava, Shyamprasad Karagadde
Department of Mechanical Engineering, Indian Institute of Technology Bombay
Mumbai, India 400076

S. Scale analysis

From the literature [1], the length scale for salt-finger cells (d) in binary alloy solidification, is formulated as Eq. (S.1).

$$d \sim 2\pi \left(\frac{D_S \nu}{g \frac{\Delta S}{h} \beta_S} \right)^{1/4} \tag{S.1}$$

where D_S is the compositional diffusivity (m^2/s), ν is the kinematic viscosity (m^2/s), g is the gravitational acceleration (m/s^2), h is the height of the liquid, β_S is the compositional expansion coefficient in the binary system, ΔS is the composition difference at the onset of convection (wt %) in the binary system. In the binary mixture, the scaled diameter of salt-finger or plume (d from Eq. (S.1)) is inversely proportional to $(\beta_S \Delta S)^{1/4}$, but in ternary systems, the proportionality elements are not known in cases where two different compositional expansion coefficients and composition differences are experienced during the convection. For studying such systems, a linear stability analysis has been performed to validate the present experimental studies. A stabilizing temperature gradient inhibits the onset of convection in a fluid that is subjected to a positive composition gradient in the multi-component system. The onset of instability may occur as an oscillatory motion due to the stabilizing effect of the thermal. These results are obtained from linear stability theory in two-dimensional flow and are described below.

S.1 Formulation of the problem for 2-D linear stability analysis

A fluid of height h is subjected to bottom cooling. The temperatures are given by $T = T_m$, at $y = 0$ and $T = T_m + \Delta T$ at $y = h$. The corresponding values of the compositions are $S_1 = S_{m1}$, $S_2 = S_{m2}$ at $y = 0$ and $S_1 = S_{m1} + \Delta S_1$, $S_2 = Sm_2 + \Delta S_2$ at $y = h$ (figure S1). For the linear stability analysis, temperature and composition can be divided into two parts, (i) the linear part given above and (ii) the part due to convective redistribution, and it is formulated in Eq. (S.2).

$$T_{total} = T_m + \Delta T \frac{y}{h} + T(x, y, z, t) \tag{S.2a}$$

$$S_{total1} = S_{m1} + \Delta S_1 \frac{y}{h} + S_1(x, y, z, t) \tag{S.2b}$$

$$S_{total2} = S_{m2} + \Delta S_2 \frac{y}{h} + S_2(x, y, z, t) \tag{S.2c}$$

where T is temperature, S_1, and S_2 are compositions of the first and second components, and subscript m indicates the mean value. The boundary conditions are taken to be of no flux type for heat and compositions at the top and bottom. Furthermore, this analysis is restricted to two-dimensional motions.

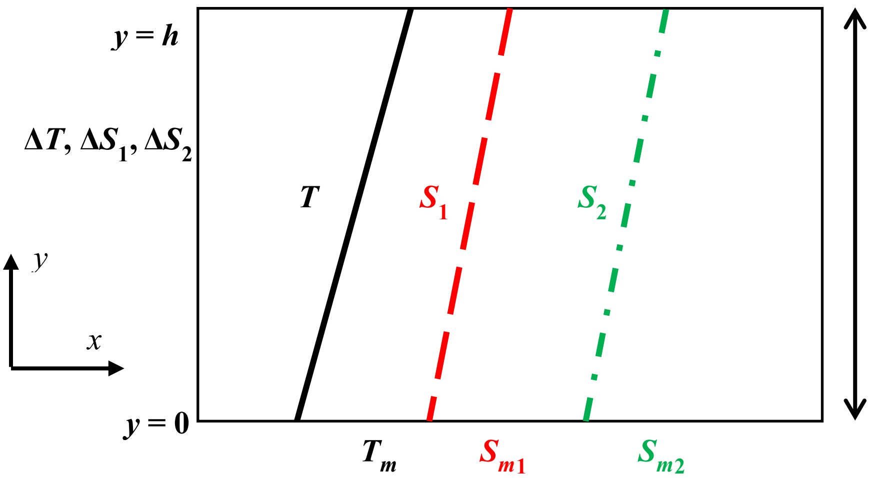

Figure S1: Schematic of the analytical problem description.

S.2 Governing equations

The governing equations of momentum, mass, temperature, and composition are used to assume that the flow is incompressible. The Boussinesq term is incorporated in momentum equations to represent natural convection arising from compositional gradients and is given by:

$$\frac{\partial}{\partial t}\mathbf{V} + \mathbf{V}.\nabla\mathbf{V} = -\frac{1}{\rho_m}\nabla p + \mathbf{g}(\alpha\Delta T - \beta_1\Delta S_1 - \beta_2\Delta S_2) + \nu\nabla^2\mathbf{V} \tag{S.3a}$$

where $\mathbf{V}$ is velocity vector ((u, v) are velocities in x and y direction), t is the time, $\mathbf{g}$ is the gravitational acceleration, p is the pressure, ν is the kinematic viscosity. The conservation of mass,

$$\frac{\partial \mathrm{u}}{\partial x}+\frac{\partial \mathrm{v}}{\partial y}=0 \quad \text{(S.3b)}$$

The conservation of heat,

$$\frac{\partial T}{\partial t}+\mathbf{V}.\nabla T=\kappa\nabla^2 T \quad \text{(S.3c)}$$

The conservation of compositions of the two species is given by,

$$\frac{\partial S_1}{\partial t}+\mathbf{V}.\nabla S_1=\kappa_{S_1}\nabla^2 S_1 \quad \text{(S.3d)}$$

$$\frac{\partial S_2}{\partial t}+\mathbf{V}.\nabla S_2=\kappa_{S_2}\nabla^2 S_2 \quad \text{(S.3e)}$$

ρ is the body source term accounting for the thermal and compositional dependence of the liquid density, and is given by the linearized equation:

$$\rho=\rho_m(1-\alpha T+\beta_1 S_1+\beta_2 S_2) \quad \text{(S.3f)}$$

In the above equations, $\kappa,\kappa_{S_1},\kappa_{S_2}$ are the kinematic diffusivity of temperature and compositions of S_1 and S_2, respectively; ρ_m is the mean density of the system. The quantities α,β_1,β_2(> 0) can be defined as

$$\alpha=-\frac{1}{\rho}\frac{\partial\rho}{\partial T}\Big|_{S_1,S_2}\,,\quad \beta_1=\frac{1}{\rho}\frac{\partial\rho}{\partial S_1}\Big|_{T,S_2}\,,\quad \beta_2=\frac{1}{\rho}\frac{\partial\rho}{\partial S_2}\Big|_{T,S_1} \quad \text{(S.3g)}$$

S.3 Non-dimensionalization of governing equations and other parameters

Introducing the stream function (ψ) to transform the governing equation to non-dimensional forms. Stream function is defined as Eq. (S.4)

$$\mathrm{u}=\frac{\partial\psi}{\partial y},\quad \mathrm{v}=-\frac{\partial\psi}{\partial x} \quad \text{(S.4)}$$

Other parameters in governing equations are non-dimensioned as Eq. (S.5)

$$\mathbf{V}'=\mathbf{V}h/\kappa,\ t'=t\kappa/h^2,\ (x',y')=h(x,y),\ p'=p/(\rho_m\nu\kappa/h^2)\,, \quad \text{(S.5a)}$$

$$T'=T/\Delta T,\ S_1^{'}=S_1/\Delta S_1,\ S_2^{'}=S_2/\Delta S_2 \quad \text{(S.5b)}$$

From the Eq. (S.4 and S.5), the non-dimensional governing equation can be written as Eq. (S.6). For the simplicity $\mathbf{V}',t',x',y',T',S_1^{'},S_2^{'}$ is written as $\mathbf{V},t,x,y,T,S_1,S_2$. Non-dimensional momentum (which is also known as vorticity equation), temperature, and compositions equations, and other parameters are:

$$\left(\frac{1}{\sigma}\frac{\partial}{\partial t}-\nabla^2\right)\nabla^2\psi=-Ra_T\frac{\partial T}{\partial x}+Ra_{S_1}\frac{\partial S_1}{\partial x}+Ra_{S_2}\frac{\partial S_2}{\partial x}+\frac{1}{\sigma}\left(\frac{\partial\psi}{\partial x}\frac{\partial\nabla^2\psi}{\partial z}-\frac{\partial\psi}{\partial z}\frac{\partial\nabla^2\psi}{\partial x}\right) \quad \text{(S.6a)}$$

$$\left(\frac{\partial}{\partial t}-\nabla^2\right)T=-\frac{\partial\psi}{\partial x}+\left(\frac{\partial\psi}{\partial x}\frac{\partial T}{\partial z}-\frac{\partial\psi}{\partial z}\frac{\partial T}{\partial x}\right) \quad \text{(S.6b)}$$

$$\left(\frac{\partial}{\partial t}-\tau_1\nabla^2\right)S_1=-\frac{\partial\psi}{\partial x}+\left(\frac{\partial\psi}{\partial x}\frac{\partial S_1}{\partial z}-\frac{\partial\psi}{\partial z}\frac{\partial S_1}{\partial x}\right) \quad \text{(S.6c)}$$

$$\left(\frac{\partial}{\partial t}-\tau_2\nabla^2\right)S_2=-\frac{\partial\psi}{\partial x}+\left(\frac{\partial\psi}{\partial x}\frac{\partial S_2}{\partial z}-\frac{\partial\psi}{\partial z}\frac{\partial S_2}{\partial x}\right) \quad \text{(S.6d)}$$

where Non-dimensional parameters

Prandtl number $\sigma=\nu/\kappa$

Lewis number $\tau_1=\kappa_{S_1}/\kappa$, $\tau_2=\kappa_{S_2}/\kappa$

Thermal Rayleigh number $Ra_T=\dfrac{g\alpha\Delta Th^3}{\nu\kappa}$ (S.6e)

Compositional Rayleigh number $Ra_{S_1}=\dfrac{g\beta_1\Delta S_1h^3}{\nu\kappa}$, $Ra_{S_2}=\dfrac{g\beta_2\Delta S_2h^3}{\nu\kappa}$

The boundary condition in non-dimensional form formulates in Eq. (S.7)

$$\psi,\frac{\partial^2\psi}{\partial y^2}=T=S_1=S_2=0 \text{ at } y=0,\,1 \quad \text{(S.7)}$$

S.4 Linear stability analysis

The rightmost bracketed term is neglected for the linear stability analysis from Eqs. S.6(a-d). From boundary conditions (Eq. (S.7)), the solution of ψ,T,S_1,S_2 from the non-dimensional governing equations can be written as

$$\begin{aligned}\psi&=A_1e^{qt}\sin(\pi ax)\sin(n\pi y)\\ T&=A_2e^{qt}\cos(\pi ax)\sin(n\pi y)\\ S_1&=A_3e^{qt}\cos(\pi ax)\sin(n\pi y)\\ S_2&=A_4e^{qt}\cos(\pi ax)\sin(n\pi y)\end{aligned} \quad \text{(S.8)}$$

where πa and πn are wavenumbers in the horizontal and vertical direction, and q is the perturbation rate coefficient. The perturbation rate coefficient (q) can be a complex number in which the real part stands for the perturbation growth rate, and the imaginary part stands for the oscillating time behaviour of perturbation. Substituting Eq. (S.8) in Eq. (S.6 and S.7), we get a characteristic equation (Eq. (S.9)) that provides a condition to solve these equations.

$$\frac{k^2}{\sigma}(q+\sigma k^2) = -\frac{Ra_T \pi^2 a^2}{q+k^2} + \frac{Ra_{S_1}\pi^2 a^2}{q+\tau_1 k^2} + \frac{Ra_{S_2}\pi^2 a^2}{q+\tau_2 k^2} \tag{S.9}$$

where $k^2 = \pi^2(a^2+n^2)$. Using marginal stability analysis at the initiation of convection and in the absence of thermal convection, Eq. (S.9) can be simplified as Eq. (S.10)

$$\tau_2 Ra_{S_1} + \tau_1 Ra_{S_2} = \frac{\tau_1 \tau_2 k^6}{\pi^2 a^2} \tag{S.10}$$

Eq. (S.10) can be interpreted in two ways based on Lewis number in the ternary system: Case I: where Lewis numbers ($\tau_1 \neq \tau_2$) are different where these relations reach a minimum at a^2 = 0.5 and n^2 = 1 in Eq. (S.10) and it is simplified as Eq. (S.11).

$$\frac{gh^3}{\nu}\left(\frac{\beta_1 \Delta S_1}{\kappa_{S_1}} + \frac{\beta_2 \Delta S_2}{\kappa_{S_2}}\right) = 657 \tag{S.11}$$

From this, it can be concluded that the effective compositional Rayleigh number in the ternary system is proportional to $\left(\frac{\beta_1 \Delta S_1}{\kappa_{S_1}} + \frac{\beta_2 \Delta S_2}{\kappa_{S_2}}\right)$.

Case II: Both components (S_1, S_2) have the same Lewis number ($\tau_1 = \tau_2 = \tau$), and this can be written as Eq. (S.12).

$$\begin{gathered} Ra_{S_1} + Ra_{S_2} = 6.75\tau\pi^4 \\ \frac{gh^3}{\nu}\left(\frac{\beta_1 \Delta S_1 + \beta_2 \Delta S_2}{\kappa_S}\right) = 657 \end{gathered} \tag{S.12}$$

This analysis predicts that the effective compositional Rayleigh number in the ternary system is proportional to $\frac{(\beta_1 \Delta S_1 + \beta_2 \Delta S_2)}{\kappa_S}$.

In the ternary system, the scale of salt-finger can hence be evaluated, and this can be formulated as Eq. (S.13, and S.14) with the help of Eq. (S.1, S.11, and S.12).

Case (i) Lewis numbers ($\tau_1 \neq \tau_2$) are different

$$d \sim 2\pi \left(\frac{\nu h}{g} \left(\frac{\kappa_{S_1}}{\beta_1 \Delta S_1} + \frac{\kappa_{S_2}}{\beta_2 \Delta S_2} \right) \right)^{1/4} \tag{S.13}$$

Case (ii) Lewis numbers ($\tau_1 = \tau_2$) are the same

$$d \sim 2\pi \left(\frac{\nu h}{g} \frac{\kappa_S}{\beta_1 \Delta S_1 + \beta_2 \Delta S_2} \right)^{1/4} \tag{S.14}$$

If the compositional expansion coefficient of the first component is very large concerning that of the other ($\beta_1 >> \beta_2$), then the diameter of the salt-finger can be estimated to be only driven by the second component.

References

[1] V. Kumar, A. Srivastava, S. Karagadde, Compositional dependency of double-diffusive layers during binary alloy solidification: Full-field measurements and quantification, Phys. Fluids. 30 (2018) 113603. https://doi.org/10.1063/1.5049135.