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A SIMPLE MODEL FOR A MUSH

YOUNG-KYUN YANG

ABSTRACT. We have derived a simple ODE system for the mush by assuming that the temperature T, the solid fraction ϕ and the vertical component w of the velocity, depend on z only. Analytical solutions of the system have presented in case of $w << 1$ and $\phi << 1$.

1. Introduction

When a multi-component liquid is cooled and solidified, commonly, the solid phase advances from the cold boundary into the liquid as a branching forest of dendritic crystals. This creates a region of mixed solid and liquid phases, referred to as a mushy zone, in which the solid forms a rigidly connected framework with the liquid occuring in the intercrystalline gaps.

We consider a container having a liquid alloy or solution whose composition is less than the eutectic composition ξ_E. Suppose that a freezing interface advances steadily upward at speed U as the system is cooled from the bottom. Let T_∞ and ξ_∞ be the temperature and the composition in the far field of the liquid. Then the non-convecting liquid region admits solutions $T = T_\infty + (T_i - T_\infty)e^{-(U/\kappa)z}$ and $\xi = \xi_\infty + (\xi_i - \xi_\infty)e^{-(U/D_o)z}$, where $z = 0$ represents the location of the freezing interface in a frame moving upward with the speed U. T_i is the temperature of the interface and composition ξ_i is the corresponding liquidus composition interface. κ and D_o are the thermal and compositional diffusivity in the liquid, respectively.

The solutions show that the temperature and the composition change noticeably over a distance κ/U and D_o/U, respectively, measured from

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the interface. In general, $\kappa >> D_o$, therefore the composition ahead of the interface relaxes more rapidly from ξ_i to ξ_∞ than does the temperature from T_i to T_∞. Hence, the (ξ, T) path on the phase diagram in the liquid is first a rapid change of ξ from ξ_i to ξ_∞ and then a rapid change of T from T_i to T_∞. As we see, due to this effect, the liquid ahead of the interface has a temperature less than its corresponding liquidus: it is 'constitutionally supercooled'. Constitutional supercooling occurs when the rate of solidification per unit area exceeds a critical value. The system avoids this contradictory state by increasing the area of the surface on which freezing occurs. This is manifest as a mophological instability (Langer [8]) which results in a convoluted freezing interface. When the rate of freezing is far above the critical value, the convolution of the interface becomes extreme, with the surface effectively filling a finite volume, which we call a mush.

We treat the mush as a new single continuum phase. The temperature and the composition of solute in the interstitial fluid are approximately uniform on the scale that is small compared with the macroscopic dimensions of the system but large compared with the pore size between the crystals of the matrix. We assume that the mush is in a complete equilibrium at all times. In other words, the time-scales of melting and freezing processes are negligibly short compared with those of principal interest in studying the evolution of the mushy zone. A system in motion is clearly not in thermodynamic equilibrium, but if the motion is sufficiently slow a state of local thermodynamic equilibrium will prevail.

Thus the temperature and composition are required to satisfy the liquidus relation $T = T_r(\xi)$, which we approximate here by the linear expression

$$T = T_r - \Gamma(\xi - \xi_\infty) \tag{1.1}$$

where ξ_∞ is a reference value of the composition of the liquid, T_r is the liquidus temperature of liquid having composition ξ_∞, the liquidus slope Γ is a positive constant. The linear liquidus relationship is a common metallurgical approximation which is mathematically convenient and apparently leads to good practical results.

2. Governing equations and boundary conditions

2.1. General equations and conditions

We introduce a set of governing equations and boundary conditions for a mush that have been proposed by Hills *etc.* [5], based on principles of diffusive mixture theory. In this study, the density is treated as a constant everywhere except in the buoyancy term and the solid phase is constrained to move in rigid-body motion. The conservation of total mass requires that

$$\nabla \cdot \mathbf{w} = 0 \tag{2.1}$$

where $\mathbf{w}$ the mass flux of interdendritic fluid relative to the solid phase. We assume that the thermal conductivity, the specific heat per unit volume and the latent heat of solidification per unit volume, are constant and independent of phases. Also, we assume that the solutal diffusivity in the liquid is constant and the diffusivity in the solid phase is neglected.

The equations describing conservation of a constitutent in the liquid phase and energy in both phases can be written as

$$\frac{D_s\xi}{Dt} + \mathbf{w} \cdot \nabla\xi = \frac{D_s(\phi\xi)}{Dt} + D_o\nabla \cdot ((1-\phi)\nabla\xi) \tag{2.2}$$

$$\rho_r c_p(\frac{D_sT}{Dt} + \mathbf{w} \cdot \nabla T) = k\nabla^2 T + \rho_r L\frac{D_s\phi}{Dt} \tag{2.3}$$

where D_o is the material diffusivity in the liquid phase, L is the latent heat, c_p is the specific heat, k is the thermal conductivity, ρ_r is a reference density, ϕ is the mass fraction of solid, ξ is the mass fraction of light constitutent of the liquidus, and $D_s/Dt = \partial/\partial t + \mathbf{u}^s \cdot \nabla$ is the material derivative following the solid phase. The first term on the right hand side of the equation (2.2) represents the increase (or decrease) of composition of the liquid due to freezing (or melting) of the solid phase, while the second is the Fickian diffusive term which is commonly negligibly small. The first term on the right hand side of the equation (2.3), represents the thermal diffusion, and the second is the release (absorption) of latent heat of fusion as solid phase freezes (melts).

The solid phase is assumed to be rigidly attached to a substrate so that only the motion of the liquid phase is of concern. The percolation of the liquid phase relative to the solid is assumed to be governed by Darcy's law:

$$\frac{\eta \mathbf{w}}{\gamma(1-\phi)^2} = -\nabla p - (\rho^l - \rho_r) g \mathbf{z} \tag{2.4}$$

where η is the dynamic viscosity of the liquid, p is the dynamic pressure *i.e.* the hydrostatic pressure field subtracted, ρ^l is mass of liquid per unit volume of mixture, g is the gravity, $\mathbf{z}$ is the unit upward vector, and γ is the permeability of the mush. Worster ([16]) used

$$\gamma = \gamma_o (1-\phi)^3, \tag{2.5}$$

which is suggested by the form of the Kozeny equation. We note that the Kozeny-Carmen equation $\gamma = (1-\phi)^3 d^2/180\phi^2$ (d is the base diameter of a slender cone approximating the dendrite.) used by Chen & Chen ([1]), in which $\gamma \to \infty$ as $\phi \to 0$, is inappropriate when, as in this study, the Darcy equation is used to describe the flow in the porous medium rather than the more general Brinkman equation (Worster [16]). We use equation (2.5) for the permeability.

Since both thermal and compositional gradients exist across the mush, we must take account of both in calculating the overall density difference driving compositional convection. Thus, to determine the buoyancy forcing in (2.4), we use a linearized equation of state

$$\rho^l = \rho_r [1 - \alpha(T - T_r) - \beta(\xi - \xi_r)] \tag{2.6}$$

where α and β, are coefficients of thermal and compositional expansion, assumed constant. Note that β is positive since ξ is the mass fraction of light constitutent. Within the mush, this relationship can be written as

$$\rho^l = \rho_r [1 + (\frac{\beta}{\Gamma} - \alpha)(T - T_r)] \tag{2.7}$$

by taking the liquidus relationship (1.1) into account. Note that $(\beta/\Gamma) - \alpha$ is usually positive since $\beta/(\alpha\Gamma)$ is typically much larger than unity. Now, if we substitute equation (2.7) into (2.4), we get

$$\frac{\eta \mathbf{w}}{\gamma(1-\phi)^2} = -\nabla p - \rho_r (\frac{\beta}{\Gamma} - \alpha)(T - T_r) g \mathbf{z}. \tag{2.8}$$

Equations (1.1), (2.1), (2.2), (2.3) and (2.8) constitute a full set of governing equations for the variables T, p, ξ, ϕ and $\mathbf{w}$ within the mush.

Three interfacial conditions that express conservation of mass, energy and solute at both solid-mush and mush-liquid interfaces can be derived directly by integrating equations (2.1), (2.2) and (2.3) over an elementary volume enclosing (and collaping onto) each interface. These can be expressed as

$$[\mathbf{w} \cdot \mathbf{n}]_-^+ = 0, \tag{2.9}$$

$$\rho_r[-c_pT + L\phi]_-^+ V_n = [(-\rho c_p T(\mathbf{u}^s + \mathbf{w}) + \rho_r L\phi\mathbf{u}^s + k\nabla T) \cdot \mathbf{n}]_-^+, \tag{2.10}$$

$$[(1-\phi)\xi]_-^+ V_n = [((1-\phi)\xi\mathbf{u}^l - D_o(1-\phi)\nabla\xi) \cdot \mathbf{n}]_-^+, \tag{2.11}$$

where V_n is the normal velocity of the solid-mush or mush-liquid interface, $\mathbf{n}$ is a unit vector normal to the interface and the square brackets denote the jump in the enclosed quantity across the interface. Also, we require that the pressure , temperature and liquid composition be continuous at the mush-liquid interface, *i.e.*,

$$[p]_-^+ = 0, \qquad [T]_-^+ = 0, \qquad [\xi]_-^+ = 0. \tag{2.12}$$

Finally, we adopt a configuration of marginal thermodynamic equilibrium suggested by Worster ([14]), which is achieved if the temperature gradient on the liquid side of the mush-liquid interface is equal to the gradient of the local liquidus temperature. This is expressed by

$$\mathbf{n} \cdot \nabla T = -\Gamma \mathbf{n} \cdot \nabla \xi. \tag{2.13}$$

But, since we assume that the thermal conductivity is independent of phases, the marginal equilibrium condition (2.13) is equivalent to

$$\phi = 0, \tag{2.14}$$

on the mush-liquid interface (Worster [14]).

Now, the boundary conditions to be applied to the mush-liquid interface are (2.9) through (2.12) and (2.14). The boundary conditions on the solid-liquid interface, consist of (2.9), (2.10), (2.12.2) and

$$T = T_e, \tag{2.15}$$

where T_e denotes the eutectic temperature. Note that if growth is not eutectic, then the equation (2.12.2) will be used instead of (2.15). (Fowler [3]).

2.2. Steady nondimensional problem

We assume the system to be steady in a frame fixed to the mush-solid interface, which moves upward relative to the solid with a prescribed constant speed V. The liquid region has fixed temperature T_∞ and composition ξ_∞ of light constitutent as $z \to \infty$, where z measures vertical displacement in the moving frame. The temperature decreases downward, and we consider the case in which a mushy zone separates a completely solid region from a completely liquid region. In this model problem we assume that the eutectic front, at which the temperature is equal to the eutectic temperature T_e and below which the system is completely solid, can be maintained at the fixed position $z = 0$. The mush-liquid interface $z = h$ is a free boundary to be determined as part of the solution. In general $h = h(x, y)$ though, in our case, it will be assumed a constant, as suggested by the experiments of ammonium chloride solution (Roberts & Loper [11], Chen & Chen [1]). We nondimensionalize the governing equations and boundary conditions by choosing a thermal length scale κ/V and thermal time scale κ/V^2, where κ is the thermal diffusivity $\kappa = k/\rho_r c_p$. Specifically, put $\mathbf{x} = (\kappa/V)\mathbf{x}^*$, $\mathbf{w} = V\mathbf{w}^*$, $p = \kappa\eta/\gamma_o p^*$, $\gamma = \gamma_o\gamma^*$, $T - T_r = (T_r - T_e)T^*$, $\xi - \xi_\infty = (\xi_e - \xi_\infty)\xi^*$, where T_r is the liquidus temperature of ξ_∞. Dropping the asterisks, (2.1), (2.2) and (2.8) become

$$\nabla \cdot \mathbf{w} = 0, \tag{2.16}$$

$$\mathbf{w}.\nabla\xi = \frac{\partial\xi}{\partial z} - \frac{\partial(\phi\xi)}{\partial z} - C\frac{\partial\phi}{\partial z}, \tag{2.17}$$

$$\mathbf{w} \cdot \nabla T = \nabla^2 T + \frac{\partial T}{\partial z} - S\frac{\partial\phi}{\partial z}, \tag{2.18}$$

$$\frac{\mathbf{w}}{\gamma(\phi)(1-\phi)^2} + \nabla p + R_a T\mathbf{z} = 0, \tag{2.19}$$

$$T = -\xi. \tag{2.20}$$

The parameters are a Stefan number $S = L/c_p(T_r - T_e)$, which represents the ratio of the latent heat needed to melt the solid and the heat needed to warm the solid from its eutectic temperature to the reference temperature T_r, the ratio of composition $C = \xi_\infty/(\xi_e - \xi_\infty)$, which denotes the compositional contrast between solid and liquid phases compared to the typical variations of concentration within the liquid (Worster [15]), and a Rayleigh number $R_a = \gamma_o \rho_r (\beta - \alpha\Gamma) g (T_r - T_e)/V\eta\Gamma$, which will act to drive buoyancy induced convection in the mush if it is large enough. Note that very large Lewis number $Le = \kappa/D_o$ is assumed in equation (2.18). Boundary conditions are:

(i) on the liquid-mush interface, $z = h(x, y)$

$$\phi = 0, \qquad [\mathbf{w} \cdot \mathbf{z}]^+_- = 0, \qquad [p]^+_- = 0, \qquad [\xi]^+_- = 0,$$

$$[T]^+_- = 0, \qquad [\frac{\partial T}{\partial z}]^+_- = 0, \qquad [\frac{\partial \xi}{\partial z}]^+_- = 0.$$

(ii) on the solid-mush interface, $z = 0$

$$\mathbf{w} \cdot \mathbf{z} = 0, \qquad T = -1, \qquad \xi = 1.$$

3. A simple model for a mush

We derive a simple ODE system, present analytical solution of the model in case of $w << 1$ and $\phi << 1$. The knowledge of ϕ provides us the structure of the mush. We obtain the expression of the constant thickness h_0 of the mush.

3.1. Derivation of a simple ode system

We assume that T, ϕ and w depend on z only. Then we derive a ODE system consisting of three equations.

If $T = T(z)$ and let $-w$ be the vertical component of the velocity, from (2.17) and (2.18), we have

$$-wT' = T' - (\phi T)' + C\phi', \tag{3.1}$$

$$-wT' = T'' + T' - S\phi', \tag{3.2}$$

where the liquidus relation (2.20) has been used in (3.1) and the prime $'$ denotes the derivative with respect to z. From (3.1) and (3.2), we get

$$T' = (C + S - T)\phi + H, \tag{3.3}$$

where $H = T'(h_0)$ measures the amount of superheat. (3.3) If we solve (3.1) for ϕ', we obtain the second of the set of three equations

$$\phi' = \frac{T'}{T-C}(1+w-\phi). \tag{3.4}$$

Within the mush, we let $\mathbf{w} = \mathbf{w}_H - w(z)\hat{\mathbf{z}}$, where $\mathbf{w}_H$ denotes the horizontal velocity vector. Then we have from (2.16) and (2.19)

$$\nabla_H \cdot \mathbf{w}_H - w' = 0, \tag{3.5}$$

$$\frac{\mathbf{w}_H}{\gamma(\phi)(1-\phi)^2} + \nabla_H p = 0, \tag{3.6}$$

$$\frac{-w}{\gamma(\phi)(1-\phi)^2} + \frac{\partial p}{\partial z} + R_a T = 0. \tag{3.7}$$

If we take ∇_H of (3.7), we obtain

$$\nabla_H(\frac{\partial p}{\partial z}) = 0, \tag{3.8}$$

which is solved by

$$p = p_a(z) + p_b(\mathbf{x}_H). \tag{3.9}$$

Dividing (3.6) by $\gamma(\phi)(1-\phi)^2$ and differentiating with respect to z yields

$$[(\nabla_H \cdot \mathbf{w}_H - w')/\gamma(\phi)(1-\phi)^2]' = 0. \tag{3.10}$$

If we take $\frac{\partial}{\partial z}\nabla_H \cdot$ (3.6), (3.10) and (3.8) imply

$$(\frac{w'}{\gamma(\phi)(1-\phi)^2})' = 0. \tag{3.11}$$

Integrating (3.11) gives

$$w' = V\gamma(\phi)(1-\phi)^2, \tag{3.12}$$

where $V = w'(h_0)$. Now, the set of equations is (3.3), (3.4) and (3.12) involving variables T, ϕ and w .

The boundary conditions are

$$T(h_0) = 0, \qquad \phi(h_0) = 0, \qquad T(0) = -1, \qquad w(0) = 0. \tag{3.13}$$

Note that we use (3.12.3) to find the thickness h_0 of the mush.

3.2. Analytic solutions

The system consisting of (3.3), (3.4) and (3.12) plus boundary conditions have analytical solutions in case of $w << 1$ and $\phi << 1$.

We assume that $w << 1$. Then from (3.3) and (3.4), we have

$$\phi = \frac{T}{T - C}, \tag{3.14}$$

where the conditions $T_h = 0$ and $\phi_h = 0$ were used.

(3.15)

$$z = \frac{1}{2} \ln \frac{1 + A + B}{T^2 - AT + B} + (\frac{2C - A}{2\sqrt{D}}) \ln \frac{A + 2B + \sqrt{D} - T(2 + A - \sqrt{D})}{A + 2B - \sqrt{D} - T(2 + A + \sqrt{D})},$$

where

$$A = C + S + H, \qquad B = CH, \qquad D = A^2 - 4B.$$

The solution (3.14) for ϕ reveals that the solid fraction in the mush decreases when the temperature increases, and shows that the ratio of composition C affects the distribution of solid in the mush. The depth of the mush is obtained by setting $z = h_0$, $T = 0$ in (3.15). Similar expressions to (3.14) and (3.15) are given by Hills *al.*([5]), Fowler([3]) and Worster([15]).

The case $C >> 1$ and $\phi << 1$ is typical of the experiments using aqueous solutions of ammonium chloride. This limit yields explicit functions w, ϕ and T of z. The value of h_0 depends on V and H if $S << C$. This means that the thickness of the mush is determined dominantly by the strength of convection.

If $C >> 1$ and $\phi << 1$, from (3.3), (3.4) and (3.12), we obtain

$$w = Vz, \tag{3.16}$$

$$\phi = \frac{H}{C + S}[e^{\frac{(C+S)V}{2C}[(h-\frac{1}{V})^2-(z-\frac{1}{V})^2]} - 1], \tag{3.17}$$

where $\phi_h = 0$ was used,

$$T + 1 = \sqrt{\frac{\pi C}{2(C + S)V}} H e^{\zeta^2(h)}[\mathbf{erf}(\zeta) - \mathbf{erf}(\zeta(0))], \tag{3.18}$$

where

$$\zeta = \sqrt{\frac{(C+S)V}{2C}}(z + \frac{1}{V}), \qquad \mathbf{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2}\, dt.$$

Note that the thickness h_0 of the mush is obtained by setting $z = h_0$, $T = 0$ in (3.18), and it is independent of C if $S << C$, *i.e.*,

$$(3.19) \quad 1 = \sqrt{\frac{\pi}{2V}} H e^{\zeta^2(h_0)} [\mathbf{erf}(\zeta) - \mathbf{erf}(\zeta(0))], \qquad \zeta = \sqrt{\frac{V}{2}}(z + \frac{1}{V}).$$

We solve (3.19) numerically for h_0. We see that h_0 decreases as the strength of convection V increases. This confirms the fact that convection increases the heat transfer from the liquid above the mush.

References

[1] Chen, C. F. & Chen. F., *Experimental study of directional solidification of aqueous ammonium chloride solution*, J. Fluid Mech. **227** (1991), 567-586.

[2] Copley, S. M., Giamei, A. F., Johnson, S. M. & HORNBECKER, M. F., *The origin of freckles in unidirectionally solidified castings*, Metall. Trans. **1** (1970), 2193-2204.

[3] Fowler, A. C., *The formation of freckles in binary alloys*, IMA J. Appl. Math. **35** (1985), 159-174.

[4] Hellawell, A., Sarazin, J.R. & Steube, R.S., *Channel convection in partly solidified systems*, Phil. Trans. R. Soc. Lond. **A 345** (1993), 507-544.

[5] Hills, R. N., Loper, D. E. & Roberts, P. H., *A thermodynamically consistent model of a mushy zone*, Q.J. Appl. Maths **36** (1983), 505-539.

[6] Huppert, H. E., *The fluid mechanics of solidification*, J. Fluid Mech. **212** (1990), 209-240.

[7] Huppert, H. E. & Worster, M. G., *Dynamic solidification of a binary alloy*, Nature **314** (1985), 703-707.

[8] Langer, J. S., *Instabilities and pattern formation in crystal growth*, Rev. Mod. Phys. **52** (1980), 1-28.

[9] Loper, D. E., *Structure of the inner core boundary*, Gephys. Astrophys. Fluid Dyn. **25** (1983), 139-155.

[10] ——, *Structure and dynamics of partially solidified systems*, Martinus Nijhoff, 1983

[11] Roberts, P. H. & Loper, D. E., *Towards a theory of the structure and evolution of a dendrite layer*, In Stellar and Planetary Magnetism (Soward, A. M., Ed.) 1983, 329-349.

[12] Tait, S. & Jaupart, C., *Compositional convection in viscous melts*, Nature **338** (1989), 571-574.

[13] ——, *Compositional convection in a reactive crystalline mush and the evolution of porosity*, J. Geophys. Res. **97**(B5) (1991), 6735-6756.
[14] Worster, M. G., *Solidification of an alloy from a cooled boundary*, J. Fluid Mech. **167** (1986), 481-501.
[15] ——, it Natural convection in a mushy layer, J. Fluid Mech. **224** (1991), 335-359.
[16] ——, it Instabilities of the liquid and mushy regions during solidification of alloys, J. Fluid Mech. **237** (1992), 335-359.

YOUNG-KYUN YANG, DEPARTMENT OF MATHEMATICS, YONSEI UNIVERSITY, SEOUL 120-749, KOREA