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AN ANALYSIS OF THE CHIMNEY WALL

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Abstract. As seen from the ammonium chloride experiment (Chen & Chen [1], Roberts & Loper [11]), the interface near chimneys has an up-rising shape and we observe thickening of mush next to chimney. We analyze the thermal boundary layer around chimney that forms as the mush is cooled locally by the fluid rising through the chimney. We obtain solutions of the temperature, the solid fraction, and the pressure in the chimney wall. Also, our expression of the pressure shows that the fluid flow can require a huge pressure in order to pass through the chimney wall if its permeability is very small. We present a simple analytic description of the up-rising shape near the exit of the chimney, due to the fact that the comparatively solute (*i.e.* NH_4Cl in the case of the ammonium chloride experiment)-rich fluid near the chimney tends to crystallize as it is chilled by the rising jet of cold fluid in the chimney.

1. Introduction

Copley et al.(1970) reported experiments in which they had cooled and crystallized from below aqueous solutions of ammonium chloride. This particular salt was chosen because its crystal habit is similar to that of many metallic alloys. The authors found that convection of buoyant fluid from the interstices of the mushy layer, which formed as crystals of ammonium chloride grew at the base of the container, took the form of narrow, vertical plumes rising through crystal-free vents or 'chimneys' in the dendritic matrix. They suggested that these convectively formed chimneys are the cause of the 'freckles' that are often observed in castings of steel and binary alloy systems such as aluminum-copper, lead-tin

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and nickel-aluminum. Freckles are imperfections that interrupt the uniformity of the microstructure of a casting, causing areas of mechanical weakness.

The laboratory experiment is easy to perform and makes a simple and attractive fluid-dynamical demonstration. A warm aqueous ammonium chloride solution (for example, 28wt%NH_4Cl) of composition more concentrated than the eutectic value (20wt%) is placed in a suitable container (a glass beaker will do) and cooled from below, for example by placing the glass beaker on a bed of ice. After a short while the bottom of the beaker is completely covered with small dendritic crystals of ammonium chloride and the thickness of the layer gradually increases with time. It appears that a planar solid-liquid interface is highly (morphologically) unstable, and that the actual interface is a highly irregular surface, which takes the form of small dendrite arms occupying a zone of finite thickness, often called the mushy zone. The reason for this fact is that a planar interface leads to 'constitutional' supercooling, where the liquid ahead of the interface has a temperature below the liquidus, despite being above the interfacial temperature, because of the dependence of the liquidus (freezing) temperature on solute concentration. The common occurrence of dendritic or mushy zones is due to the fact that compositional diffusivities are invariably much smaller than thermal diffusivities, so that the thickness of the region over which the concentration changes in the liquid is much thinner than the corresponding region over which the temperature changes. In the early stages, finger-like convection starts in the fluid region just above the mush. With increased time, plumes with associated chimneys are visible. During an extended period these coexist with the fingers. The finger convection become progressively weaker and chimneys are eventually the only sites of upwelling. Downward flow in the mush causes solidification and upward flow promotes dissolution. The nonlinear interaction of dissolution and convection leads to the formation of chimneys (Loper [10]). The NH_4Cl solution above the mush remains continually undersaturated. The circulation above the mush consists of upward flow in isolated buoyant plumes which is compensated by a downward return flow of the undersaturated solution. This liquid slowly migrates towards the layer of crystals and flows down to it. Because the flow through the crystals takes place in a decreasing temperature field, the aqueous solution becomes saturated and NH_4Cl exsolves onto existing NH_4Cl crystals, which exhibit secondary and tertiary branching. The NH_4Cl-depleted solution from a wide area within the mush flows to a central point and the less-dense return flow takes place through a few isolated chimneys. The upward flow in the plume induces motion of the bulk fluid toward the plume itself. The comparatively solute-rich fluid, meeting the cold plume, tends to crystallize around the chimney. As a result, there is a buildup of crystals around each chimney exit, like a mini-volcano. With time some chimneys become inactive and the overall intensity of the convective motion decreases as the thickness of the mushy layer increases. (Huppert [7]). The purpose of this study is to present that the fluid flow can require a huge pressure if the permeability in the chimney wall is small, and is to analyze the up-rising top near the exit of the chimney by using the temperature expression in the chimney.

2. An axisymmetric model for a mush

We consider an axisymmetric model of a mush-chimney system containing only one chimney. 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. We nondimensionalize the governing equations by choosing a thermal length scale κ/V and thermal time scale κ/V^2, where κ is the thermal diffusivity $\kappa = k/\rho_r c_p$, c_p is the specific heat, k is the thermal conductivity, and ρ_r is a reference density. (Yang [16]). 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 γ_o is a reference value of the permeability of the mush, η is the dynamic viscosity of the liquid, T_r is the liquidus temperature of ξ_∞ and ξ_e is the eutectic composition of light constitutent. Dropping the asterisks, conservation of total mass, conservation of a constitutent in the liquid phase and energy, the liquid momentum equation and the liquid relation, respectively, become

$$\nabla \cdot \mathbf{w} = 0, \tag{1}$$

$$\mathbf{w}.\nabla\xi = \frac{\partial \xi}{\partial z} - \frac{\partial(\phi\xi)}{\partial z} - C\frac{\partial \phi}{\partial z}, \tag{2}$$

$$\mathbf{w} \cdot \nabla T = \nabla^2 T + \frac{\partial T}{\partial z} - S\frac{\partial \phi}{\partial z}, \tag{3}$$

$$\frac{\mathbf{w}}{\gamma(\phi)(1-\phi)^2} + \nabla p + R_a T \mathbf{z} = 0, \tag{4}$$

$$T = -\xi. \tag{5}$$

Let (r, θ, z) be cylindrical coordinates with z upwards. We assume within the main body of the mush that the vertical velocity w, the temperature T, the mass fraction of light constitutent ξ and the mass fraction of solid ϕ, depend on z only. Then the governing equations for the mush are

$$\frac{\partial(ru_m(r,z))}{\partial r} - \frac{\partial(rw_m(z))}{\partial z} = 0 \tag{6}$$

$$-w_m(z)T_m'(z) = T_m'(z) - (\phi_m(z)T_m(z))' + C\phi_m'(z) \tag{7}$$

$$-w_m(z)T_m'(z) = T_m''(z) + T_m'(z) - S\phi_m'(z) \tag{8}$$

$$\frac{u_m(r,z)}{F(\phi_m(z))} + \frac{\partial p_m(r,z)}{\partial r} = 0, \tag{9}$$

$$\frac{-w_m(z)}{F(\phi_m(z))} + \frac{\partial p_m(r,z)}{\partial z} + R_a T_m(z) = 0, \tag{10}$$

$$T_m(z) = -\xi_m(z), \tag{11}$$

where the prime $'$ denotes the derivative with respect to z, and $F(\phi_m) = (1-\phi_m)^5$ for Worster's choice [15]. 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, where L is the latent heat, Γ is the liquidus slope, g is the gravity, α and β, are constant coefficients of thermal and compositional expansion. Note that very large Lewis number $Le = \kappa/D_o$ is assumed in equation (3), where D_o is the compositional diffusivity in the liquid. ¿From the above equations, we obtain the set of equations involving variables $T_m(z)$, $\phi_m(z)$ and $w_m(z)$:

$$T'_m = (C + S - T_m)\phi_m + H, \tag{12}$$

$$\phi'_m = \frac{T'_m}{T_m - C}(1 + w_m - \phi_m), \tag{13}$$

$$w'_m = WF(\phi_m), \tag{14}$$

$$p_m(r,z) = p_a(z) + p_b(r) \tag{15}$$

with the boundary conditions

$$T_m(h_0) = 0, \qquad \phi_m(h_0) = 0, \qquad T_m(0) = -1, \qquad w_m(0) = 0, \tag{16}$$

where h_0 is a constant mush-liquid interface away from the chimney wall, as suggested by the experiments of ammonium chloride solution (Roberts & Loper [11], Chen & Chen [1]), $H = T'(h_0)$ measures the amount of superheat and $W = w'(h_0)$.

3. An analysis of chimney wall

In this section, we assume that $C - T_m >> S$, and the thickness ϵ of chimney wall is small as seen from the ammonium chloride experiments. We find the temperature and the solid fraction in the chimney wall. Furthermore, we obtain an analytical solution of pressure in the chimney wall by assuming that the solid fraction away from the chimney wall is much less than 1 and the radius of the chimney is constant.

We let

$$u_{cw} = u_m(r,z), \qquad w_{cw} = w_m(z), \tag{17}$$

$$T_{cw} = T_m(z) + T_1(r,z), \qquad \xi_{cw} = \xi_m(z) + \xi_1(r,z), \tag{18}$$

$$\phi_{cw} = \phi_m(z) + \phi_1(r,z), \qquad p_{cw} = p_a(z) + p_1(r,z), \tag{19}$$

where subscripts 'cw' represent the chimney wall. Note that $p_1(r,z)$ contains the horizontal pressure $p_b(r)$ of the mush. Integrating the continuity equation yields

$$u_{cw}(r,z) = \frac{-R^2}{2r} w'_m(z), \tag{20}$$

where R is the radius of the cylindrical catchment of one chimney. Note that we neglected ϵ^2 compared to R^2 in (20), and $u_{cw} = O(1/\epsilon)$.

If we substitute (17)~(19) into (2) and (3), we have by using the liquidus relation

$$u_{cw} \frac{\partial T_1}{\partial r} = \frac{\partial}{\partial z}[(C - T_0)\phi_1], \tag{21}$$

$$u_{cw} \frac{\partial T_1}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r}(r \frac{\partial T_1}{\partial r}) - S \frac{\partial \phi_1}{\partial z}, \tag{22}$$

where we have used (7) and (8) in (21) and (22), respectively, and we neglected $w_{cw}\partial T_1/\partial z$, $\partial T_1/\partial z$, $\partial \phi_m T_1/\partial z$ compared to $u_{cw}\partial T_1/\partial r$ in (21) and $\partial^2 T_1/\partial z^2$, $\partial T_1/\partial z$ compared to $\partial(r\partial T_1/\partial r)/r\partial r$ in (22).

In order to get a simple solution, we assume that $C - T_0 >> S$. Then we may neglect the second term in (22) on the right (contribution due to the latent heat) compared to the advection term on the right. Then we have

$$\frac{-R^2}{2r} w'_m(z) \frac{\partial T_1}{\partial r} = \frac{1}{r} \frac{\partial}{\partial r}(r \frac{\partial T_1}{\partial r}), \tag{23}$$

where u_{cw} has been replaced by (20). If we integrate (23), we get

$$T_1(r,z) = T_1(a,z)(\frac{a}{r})^{2Q(z)}, \tag{24}$$

where $T_1(a,z)$ denotes the temperature on the wall of chimney, and

$$Q = \frac{R^2 w'_m(z)}{4} = \frac{WR^2(1-\phi_m)^5}{4}, \tag{25}$$

where Worster's permeability relation has been used in (25).

We want to obtain a simple analytical solution for pressure in the chimney wall, so that we assume that $\phi_m(z) << 1$ and a is constant. Then we get $w'_m(z) = W$, and $Q = R^2 W/4$ which is constant.

In order to obtain $\phi_1(r,z)$, we substitute (24) into (21), and integrate (21) with respect to z after replacing u_{cw} by (20). So, we get

$$\phi_1(r,z) = \frac{1}{(C-T_0)} \int_h^z \frac{4QT_1(a,z)}{a^2} (\frac{a}{r})^{2Q(z)+2}\, dz. \tag{26}$$

Note that ϕ_1 satisfies the boundary condition $\phi_1(r,h) = 0$ at the top of the mush. If Q is constant, we can rewrite $\phi_1(r,z)$ as

$$\phi_1(r,z) = \theta(z)(\frac{a}{r})^{2Q+2}, \tag{27}$$

where

$$\theta(z) = \frac{4Q}{a^2(C-T_0)} \int_h^z T_1(a,z)\, dz.$$

We obtain an expression of $p_1(r,z)$ by integrating the horizontal component of momentum equation (4). Let's substitute (17) and (19) into (4). Then we have

$$\frac{\partial p_1}{\partial r} = -\frac{u_{cw}}{(1-\phi_m(z)-\phi_1(r,z))^5}. \tag{28}$$

Now, if neglect $\phi_m(z)$ in (28), and replace u_{cw} and $\phi_1(r,z)$ by (20) and (27), respectively, we get

$$\frac{\partial p_1}{\partial x} = \frac{Q}{Q+1} \frac{x^{5Q+4}}{(x^{Q+1}-\theta)^5}, \qquad x = (\frac{r}{a})^2. \tag{29}$$

Integrating (29) yields

$$p_1 = \frac{Q}{Q+1}[\ln y - \frac{4\theta}{y} - \frac{3\theta^2}{y^2} - \frac{4\theta^3}{3y^3} - \frac{\theta^4}{4y^4}], \tag{30}$$

where

$$y = x^{Q+1} - \theta. \tag{31}$$

Note that since $x^{Q+1} = \theta/\phi_1$ from (27), $y = \theta(1/\phi_1 - 1)$ from (31). So, $y \to 0$ as $\phi_1 \to 1$. Therefore, $p_1 \to -\infty$ as $\phi_1 \to 1$ which shows that the fluid flow needs large pressure in order to get through the wall if the permeability of the wall is very small.

4. Shape of mush-liquid interface

We now analyze the up-rising shape of a chimney. Let's assume that on the top of the mush,

$$z = h(r) = h_0 + h_1(r). \tag{32}$$

We take Taylor series expansions of $T_{cw}(z)$ about $z = h_0$. Then we have

$$T_{cw}(r,h) = T_m(h_0) + h_1(r)T'_m(h_0) + T_1(r,h_0), \tag{33}$$

If we apply the boundary condition $T = 0$ on the top of the mush to (32), *i.e.* $T_{cw}(r,h) = 0$, $T_m(h_0) = 0$, then we obtain

$$0 = h_1(r)T'_m(h_0) + T_1(r,h_0), \tag{34}$$

If we solve (32) for $h_1(r)$, then we obtain

$$h_1(r) = -\frac{T_1(r,h_0)}{T'_m(h_0)}. \tag{35}$$

If we substitute (24) into (35), we obtain

$$h_1(r) = -\frac{T_1(a,h_0)}{H}(\frac{a}{r})^{2Q}, \tag{36}$$

where we used $T'_0(h_0) = H$ from (12).

For the ammonium chloride experiment, we estimate $Q \sim 1.0$. When $Q = 1.0$, the corresponding normalized form of $h_1(r)$ is

$$\frac{h_1(r)}{h_1(a)} = (\frac{a}{r})^2.$$

Its graph for $0 < r < a$ agrees qualitatively with the up-rising shape near the exit of the chimney and the nearly horizontal figure of the top of the mush. Note that experimental measurement of $h_1(r)$ allows us to determine both Q and $T_1(a,h_0)/H$ from (36).

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