Behavior of the Two-Phase Mushy Zone During Freeze Coating on a Continuous Moving Plate

The process of freeze coating of a binary substance on a chilled moving plate is studied theoretically with special emphasis on the behavior of the two-phase mushy zone. The flow and heat transfer in five separate regions of the system, i.e., the moving plate, the freeze coat, the two-phase packing region, the two-phase dispersed region and the molten substance region, are formulated mathematically to describe the freeze-coating process. A supplemental equation derived from a simplified phase diagram and an appropriate viscosity model are employed to complete the mathematical description of the two-phase mushy zone. The system of equations is solved by a combined analytical-numerical technique to determine the spatial variations of the solidus and liquidus fronts. Effects of seven controlling parameters, including the freeze-coat-to-wall thermal ratio, the wall subcooling parameter, the molten substance superheating parameter, the Prandtl number, the Stefan number, the equilibrium partition ratio, and the packing limit fraction, on the behavior of the two-phase mushy zone and the freeze-coating process are determined.

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Introduction

The freeze-coating process is a material manufacturing process that finds applications in electrical, chemical and thermal industries. In these applications, a chilled object is fed through a bath of molten substance. Before entering the bath, the object is subcooled below the freezing point of the substance. As the object travels through the bath, the molten substance begins to solidify on the surface of the object, thus forming a thin layer of freeze coat. Depending on which type of materials the molten substance is made of, the freeze coat may have different functions. For a polymeric substance, the freeze coat may be applied for water resistance, corrosive protection, thermal insulation, electrical insulation, or fire retardation. For a binary metallic substance, the freeze coat may have different functions. For a polymeric substance, the freeze coat may be applied for water resistance, corrosive protection, thermal insulation, electrical insulation, or fire retardation. For a binary metallic substance, the freeze coat may be applied for rust resistance, water resistance, or chemical protection.

The freeze-coating process has been investigated by many researchers. However, most of the previous studies have been restricted to the case of a pure substance in which the interface between the solidified layer and molten substance is sharp and isothermal. The early work of Kuiken [1] presented an analysis of freeze coat thickness by assuming the wall to be semi-infinite. Asymptotic solutions for small and large Prandtl numbers were obtained. Seeniraj and Bose [2] analyzed the process of freeze coating on a moving plate by assuming that the wall temperature was constant and the ambient liquid was saturated. Cheung [3,4] relaxed the assumptions made in [2] to account for the heat convection from a superheated liquid and the temperature variations within the wall. Rezaian and Poulikakos [5] examined the solidification on a moving isothermal boundary for molten substances having low and high Prandtl numbers. Mahmoud [6] studied the process of freeze coating of a polymeric substance by assuming that the freezing point of the polymeric substance is sharp and isothermal. The problem was numerically solved by a finite-element method, and the temporal variation of the freeze coat thickness was graphically presented. Cheung and Cha [7] employed a finite-difference method to study the growth and decay of a freeze coat on an axially moving cylinder. An experimental verification of the theoretical analysis was also conducted by Cheung et al. [8], using a thin copper wire to simulate the moving cylinder and water to simulate the molten substance.

Most of the previous studies of the freeze-coating process were investigated for the case where the liquid is a pure substance. For the case of a binary mixture such as a metal alloy, however, solidification would take place over a range of temperature leading to the formation of a two-phase mushy zone. Thus far, the only study of the process of freeze coating of a binary alloy was by Stevens and Poulikakos [9]. They assumed that there was no advection effect in the two-phase mixture and the heat transfer in the mushy zone was by conduction only. The effects of a variety of dimensionless parameters were identified.

In this study, freeze coating of a binary mixture on a semi-infinite moving plate is investigated, taking full account of the flow and heat transfer in the two-phase mushy zone. Equations governing five separate regions of the freeze-coating system are formulated and solved by a combined analytical-numerical technique. Seven independent controlling parameters are identified and their effects on the behavior of the two-phase mushy zone and the freeze-coating process are determined.

Problem Formulation

A schematic of the freeze-coating system under consideration is illustrated in Fig. 1. A chilled semi-infinite plate at a constant velocity, \( U_0 \), is fed continuously into an inlet slit on an adiabatic wall at \( x = 0 \) through a bath of molten binary substance. The plate is cooled at a uniform temperature, \( T_0 \), before entering the bath whereas the ambient liquid in the bath is kept constant at temperature \( T_a \). The inlet plate temperature, \( T_0 \), is lower than the solidus temperature, \( T_s \), of the binary substance whereas the bath temperature, \( T_a \), is higher than the liquidus temperature, \( T_l \), of the binary substance, (i.e., \( T_a > T_l > T_s > T_0 \)). As the plate enters the bath, a thin solidified layer or a freeze coat begins to form on the surface of the plate and continuously grows along the immersed distance. The freeze coat thickness \( \delta_1 \) and the location \( \delta_2 \) represent the isothermal contours of the solidus and liquidus temperatures, respectively.
The region \( \delta_1 \leq y \leq \delta_2 \) for which the temperature is bounded by the solidus and liquidus points, is the two-phase mushy zone. For a binary system, the mushy zone can be divided into two distinct regions (see Fig. 2): the two-phase packing region and the two-phase dispersed region, based on the value of the mixture viscosity. When the solid fraction, \( F \), in the mushy zone increases beyond a certain limit, which is called the packing limit, \( F_p \), the dendrites start locking and packing to one another and form a rigid structure trapping the liquid inside [10,11]. The viscosity in this region asymptotically approaches infinity, acting as if it were a solid. This domain of solid-like structure is the two-phase packing region. On the other hand, if \( F \) is lower than \( F_p \), the two-phase mixture is able to flow like a fluid. Based on experimental observation, the effect of force convection in the main flow can cause the dendritic structure in the mushy zone to break off [12]. The broken dendrites advected by the main flow may behave like dispersed particles suspending in the surrounding liquid. This region is referred to as the two-phase dispersed region.

To formulate the system of governing equations, a number of assumptions are made: (i) Both the molten substance and the two-phase dispersed regions behave as a Newtonian fluid. (ii) The flow is steady and laminar. Note that this is a preferred operating condition as the occurrence of transition to turbulent flow can cause a sudden decrease in the freeze coat thickness [13]. (iii) A boundary layer flow may be assumed in the molten substance and the two-phase dispersed regions. (iv) Physical properties of each phase are constant. The properties of the mushy zone are a result of the weighted average of the physical properties in each individual phase, except for the viscosity which requires a supplemental model. (v) Local thermodynamics equilibrium exists such that the solid fraction can be evaluated directly from the equilibrium phase diagram. (vi) In the two-phase dispersed region, the dispersed dendrites and the local liquid element travel at the same velocity.

As a result, there are no dispersion flux terms appearing in the momentum and energy equation. A similar approach has been employed by Voller et al. [14]. (vii) The constituent concentration in the system remains the same during the freeze-coating process. This is equivalent to assume that the effect of macrosegregation is negligible, i.e., the local composition remains constant at the initial value. Note that for the freeze-coating process, the flow is predominantly forced convection. The behavior is different than that during alloy solidification under natural convection conditions [15,16]. According to experimental observation, the higher solute concentration rejected during solidification may be washed away by the main flow in the freeze-coating process [12,17]. Nevertheless, there could be appreciable relative velocities between the liquid and the solid phases in the two-phase packing region as well as in the two-phase dispersed region, which may result in some macrosegregation [18,19]. The extent of macrosegregation, however, would not be considered in the present study, as the main focus here is the growth of the solidified layer and the behavior of the two-phase mushy zone during the freezing coating process.

With the above assumptions the governing equations for each individual region can be written along with the appropriate boundary conditions as follows:

(i) **Wall Region** \((x \geq 0 \text{ and } y \leq 0)\)

\[
\rho_v C_p \frac{\partial T}{\partial x} = k_y \frac{\partial^2 T}{\partial y^2} \tag{1}
\]

\[
x = 0: T_y = T_o \tag{2a}
\]

\[
y = 0: T_y = T_s \quad \text{and} \quad k_y \frac{\partial T_y}{\partial y} = k_s \frac{\partial T_s}{\partial y} \tag{2b}
\]

\[
y \to -\infty: T_y = T_o \tag{2c}
\]

(ii) **Freeze-Coat Region** \((x \geq 0 \text{ and } 0 \leq y \leq \delta_1)\)

\[
\rho_v C_p \frac{\partial T}{\partial x} = k_y \frac{\partial^2 T}{\partial y^2} \tag{3}
\]

\[
x = 0: \delta_1 = 0 \tag{4a}
\]

\[
y = 0: T_y = T_w \quad \text{and} \quad k_y \frac{\partial T_y}{\partial y} = k_s \frac{\partial T_s}{\partial y} \tag{4b}
\]

\[
y = \delta_1: T_y = T_1 \quad \text{and} \quad \frac{\partial T_y}{\partial y} = \frac{\partial T_{mp}}{\partial y} \tag{4c}
\]

(iii) **Two-Phase Packing Region** \((x \geq 0 \text{ and } \delta_1 \leq y \leq \delta_p)\)

\[
U_v \frac{\partial F}{\partial x} = \frac{\partial^2 T_{mp}}{\partial y^2} + \frac{\Delta H_m}{C_p_m} U_v \frac{\partial F}{\partial x} \tag{5}
\]

\[
x = 0: \delta_p = 0 \tag{6a}
\]

\[
y = \delta_1: T_{mp} = T_1 \quad \text{and} \quad \frac{\partial T_{mp}}{\partial y} = \frac{\partial T_s}{\partial y} \tag{6b}
\]

\[
y = \delta_p: T_{mp} = T_p \quad \text{and} \quad \frac{\partial T_{mp}}{\partial y} = \frac{\partial T_{md}}{\partial y} \tag{6c}
\]

(iv) **Two-Phase Dispersed Region** \((x \geq 0 \text{ and } \delta_p \leq y \leq \delta_2)\)

\[
\frac{\partial u_{md}}{\partial x} + \frac{\partial u_{md}}{\partial y} = 0 \tag{7}
\]

\[
U_{md} \frac{\partial u_{md}}{\partial x} + U_{md} \frac{\partial u_{md}}{\partial y} = \frac{\partial}{\partial y} \left( U_m \frac{\partial u_{md}}{\partial y} \right) \tag{8}
\]
\[
\begin{align*}
\frac{\partial T_{md}}{\partial x} + v_{md} \frac{\partial T_{md}}{\partial y} &= \alpha_{md} \frac{\partial^2 T_{md}}{\partial y^2} + \frac{\Delta H_m}{C_P m} \left( u_{md} \frac{\partial F}{\partial x} + v_{md} \frac{\partial F}{\partial y} \right), \\
x = 0; \delta_x = 0 \\
y = \delta_y; \quad u_{md} = u_p, \quad v_{md} = 0, \quad T_{md} = T_p \quad \text{and} \quad \frac{\partial T_{md}}{\partial y} = \frac{\partial T_{mp}}{\partial y} \quad (10a) \\
y = \delta_y; \quad u_{md} = u_l, \quad \frac{\partial u_{md}}{\partial y} = \frac{\partial u_1}{\partial y}, \quad v_{md} = v_1, \\
T_{md} = T_2 \quad \text{and} \quad \frac{\partial T_{md}}{\partial y} = \frac{\partial T_2}{\partial y}. 
\end{align*}
\]

(v) Molten Substance Region \((x \geq 0 \text{ and } y \geq \delta_y)\)

\[
\begin{align*}
\frac{\partial u_1}{\partial x} + \frac{\partial v_1}{\partial y} &= 0 \\
u_{1} \frac{\partial u_1}{\partial x} + v_1 \frac{\partial u_1}{\partial y} &= u_1 \frac{\partial^2 u_1}{\partial y^2} \\
u_{1} \frac{\partial T_1}{\partial x} + v_1 \frac{\partial T_1}{\partial y} &= \alpha_1 \frac{\partial^2 T_1}{\partial y^2} \\
x = 0; u_1 = 0 \quad \text{and} \quad T_1 = T_e \quad (14a) \\
y = \delta_y; u_1 = u_{md}, \quad v_1 = v_{md}, \quad \frac{\partial u_1}{\partial y} = \frac{\partial u_{md}}{\partial y}, \\
T_1 = T_2 \quad \text{and} \quad \frac{\partial T_1}{\partial y} = \frac{\partial T_{md}}{\partial y}. 
\end{align*}
\]

In the above formulation, the subscripts “w” refers to the wall region, “s” the freeze coat, “mp” the two-phase dispersed region, “md” the two-phase dispersed region, and “l” the molten substance region. The location \(\delta_y\) is an unknown quantity corresponding to the packing limit \(F_p\). The local temperature \(T_{mp}\) at which \(F = F_p\) is the packing limit isotherm. The right-hand side of Eqs. (1), (3), and (5) represents the advection terms due to the motion induced by the moving plate. The latent heat effect is included in the last term on the right-hand side of Eq. (5) whereas it is included in the last two terms on the right-hand side of Eq. (9). Since the two-phase packing region is forced to travel together with the plate and the freeze coat, the vertical velocity is absent and the latent heat effect is restricted to the axial direction only. In contrast, in the two-phase dispersed region, the entire flow field brings about the two-dimensional latent heat effect.

The continuous boundary conditions at the interface between each two adjacent regions result in a strong coupling of the equations governing the five distinct regions. To close the system of equations, an expression for the solid fraction must be specified in terms of the primary unknowns. Generally, the relation of the solid fraction to the local temperature of a binary substance can be graphically expressed by an equilibrium phase diagram. Following the approach of Flemings [20], the solidus and liquidus lines in the equilibrium phase diagram are assumed linear as shown in Fig. 3. Note that the equilibrium phase diagram of a binary substance can be categorized into many different regions based on the solubility of the constituents. If the value of the constituent concentration is between \(C_{min}\) and \(C_{max}\), the binary substance becomes partially soluble resulting in the formation of a eutectic structure during solidification. In the present study, the freeze-coating process is restricted to the isomorphous region given by the shaded regions in Fig. 3. An expression for the solid fraction in the isomorphous region can be derived by employing the level arm principle and the rule of similar triangle:

\[
F = \frac{T_2 - T_m}{(T_2 - T_m) + \kappa (T_m - T_1)}, \tag{15}
\]

where \(T_m\) is the local temperature of the two-phase mixture, and \(\kappa\) is the equilibrium partition ratio defined as the ratio of the slope of the liquidus line to that of the solidus line. From Eq. (15) it can be seen that the solid fraction is unity at the solidus line. It starts to decrease as the local temperature of the two-phase mushy zone is increased. When the local temperature reaches the liquidus point, the solid fraction becomes zero.

The properties of the two-phase packing and dispersed regions (represented by the subscript “m”), including the density, specific heat and thermal conductivity, are calculated by taking an average over the properties of the individual phases weighted by the local solid fraction:

\[
\rho_m = \frac{(1 - F) \rho_1 + F \rho_l}{\rho_1}, \tag{16}
\]

\[
C_{P,m} = (1 - F) C_{P_1} + F C_{P_l}, \tag{17}
\]

\[
k_m = \frac{\rho_m}{\rho_l} (1 - F) k_1 + \frac{\rho_m}{\rho_s} F k_1. \tag{18}
\]

In order to minimize the number of parameters, the density, specific heat and thermal conductivity of the freeze coat and the molten substance are treated to be the same. As a result, these properties, as well as the thermal diffusivity, of the two-phase mushy zone are identical to those for the freeze coat and molten substance.

The viscosity, on the other hand, of the two-phase dispersed region is modeled as the viscosity of dispersions (assumption iv). An expression for the viscosity of dispersions was first presented by Einstein in 1906 [21]. Einstein analytically showed that for dilute concentration of dispersed spherical particles, the intrinsic viscosity had a value of 2.5. To include the effect of high concentration and maximum packing limit, the viscosity of dispersions can be written as follows:

\[
\mu_m = \mu_0 \left( 1 - \frac{F}{F_p} \right)^{-1 - \frac{\mu F}{F_p}}. \tag{19}
\]

In terms of the dynamics viscosity, the above expression becomes

\[
u_m = \nu_0 \left( 1 - \frac{F}{F_p} \right)^{-1 - \frac{\nu F}{F_p}}, \tag{20}
\]
where $[\mu]$ is the intrinsic viscosity and has a value equal to 2.5 according to Einstein’s theory. In the above expression, $F_p$ represents the maximum packing limit of solid fraction. Equation (19), which has been experimentally verified by Krieger [10] and theoretically derived by Krieger and Daugherty [22], is known as the Krieger-Daugherty Equation. In general, the value of $F_p$ may vary from 0.5 to 0.75 depending on the structure of the two-phase mixture [23]. The local temperature $T_m = T_p$ at which $F = F_p$ can be determined by Eq. (15), and this temperature is called the packing limit isotherm. By using Eqs. (15) and (20) the system of equations for the two-phase dispersed region is mathematically closed.

Mathematical Analysis

A similarity analysis is invoked to transform the system of equations, i.e., Eqs. (1) to (20), to a set of ordinary differential equations, which can then be conveniently solved by a numerical technique. The following independent and dependent similarity variables are introduced:

(i) Wall Region

\[
\eta_w = \frac{y}{\alpha_u x} \sqrt{\frac{\sigma_s x}{U_0}} \quad -\infty \leq \eta_w \leq 0 \quad \text{and} \quad \Theta_w = \frac{T_w - T_o}{T_1 - T_o};
\]

\[
0 \leq \Theta_w \leq \Theta_w(0)
\]

(ii) Freeze-Coat Region

\[
\eta_i = \frac{y}{\delta_1}; \quad 0 \leq \eta_i \leq 1 \quad \text{and} \quad \Theta_i = \frac{\Theta - T_o}{T_1 - T_o};
\]

\[
\Theta_i(0) = \Theta_i(0) \leq \Theta_i \leq 1
\]

(iii) Two-Phase Packing Region

\[
\eta_{mp} = 1 + \frac{y - \delta_1}{\delta_2 - \delta_1}; \quad 1 \leq \eta_{mp} \leq \eta_p
\]

\[
\Theta_{mp} = 1 + \frac{T_{mp} - T_1}{T_2 - T_1}; \quad 1 \leq \Theta_{mp} \leq \Theta_p
\]

(iv) Two-Phase Dispersed Region

\[
\eta_{md} = 1 + \frac{y - \delta_1}{\delta_2 - \delta_1}; \quad \eta_p \leq \eta_{md} \leq 2,
\]

\[
f_{md} = \frac{u_{md}}{U_o}; \quad f_{md}'(2) \leq f_{md}' \leq 1
\]

\[
\Theta_{md} = 1 + \frac{T_{md} - T_1}{T_2 - T_1}; \quad \Theta_p \leq \Theta_{md} \leq 2
\]

(v) Molten Substance Region

\[
\eta_i = 1 + \frac{y}{\delta_2}; \quad 2 \leq \eta_i \leq \infty,
\]

\[
f_i' = \frac{u_1}{U_o}; \quad 0 \leq f_i' \leq f_i'(2) = f_{md}'(2)
\]

\[
\Theta_i = 2 + \frac{T_1 - T_2}{T_2 - T_1}; \quad 2 \leq \Theta_i \leq 3
\]

In the above transformation, $\eta_p$ is the similarity variable at the location where the solid fraction reaches the packing limit. The solidus and liquidus line, $\delta_1$ and $\delta_2$, respectively, and the dimensionless packing limit isotherm are given by

\[
\delta_1 = \sigma_1 \sqrt{\frac{\sigma_s x}{U_0}} \quad (26a)
\]

\[
\delta_2 = \sigma_2 \sqrt{\frac{\sigma_s x}{U_0}} \quad (26b)
\]

\[
\Theta_p = 1 + \frac{T_p - T_1}{T_2 - T_1}, \quad (26c)
\]

where $\sigma_1$ and $\sigma_2$ are the solidus and liquidus constants. Note that $\sigma_1$ and $\sigma_2$ are treated as primary unknowns in this problem. The location of the packing limit can be defined in terms of the packing limit constant $\sigma_p$ in a similar manner, i.e.,

\[
\delta_p = \sigma_p \sqrt{\frac{\sigma_s x}{U_0}} \quad (26d)
\]

Once $\sigma_1$ and $\sigma_2$ are known, $\sigma_p$ can be determined from Eqs. (23) and (26d) by setting $y = \sigma_p$ at $\eta_{mp} = \eta_{md}$. This gives

\[
\sigma_p = \left( \eta_p - 1 \right) \left( \sigma_2 - \sigma_1 \right) + \sigma_1. \quad (27)
\]

By performing the similarity transformation, the system of equations can be transformed into the following similarity forms:

(i) Wall Region

\[
\Theta_{\infty}^+ + \frac{\eta_w}{2} \Theta_w^+ = 0 \quad (28)
\]

\[
\eta_w = 0; \quad \Theta_w = \Theta_s \quad \text{and} \quad \Theta_s = \frac{R_1}{\sigma_1} \Theta_s' \quad (29a)
\]

\[
\eta_w \rightarrow -\infty; \quad \Theta_w = 0 \quad (29b)
\]

(ii) Freeze-Coat Region

\[
\Theta_i + \frac{\sigma_1}{2} \eta_i \Theta_i' = 0 \quad (30)
\]

\[
\eta_i = 0; \quad \Theta_i = \Theta_w \quad \text{and} \quad \Theta_w' = \frac{\sigma_1}{\sigma_2 - \sigma_1} R_2 \Theta_w' \quad (31a)
\]

\[
\eta_i = 1; \quad \Theta_i = 1 \quad \text{and} \quad \Theta_i' = \frac{\sigma_1}{\sigma_2 - \sigma_1} R_2 \Theta_i' \quad (31b)
\]

(iii) Two-Phase Packing Region

\[
\Theta_{mp}^+ + \frac{(\sigma_2 - \sigma_1)^2}{2} \left[ \frac{1}{1 + \frac{\kappa}{St} \left[ (2 - \Theta_{mp}) + k(\Theta_{mp} - 1) \right]^2} \right]
\]

\[
\times \left( \eta_{mp} - 1 + \frac{\sigma_1}{\sigma_2 - \sigma_1} \right) \Theta_{mp}' = 0 \quad (32)
\]

\[
\eta_{mp} = 1; \quad \Theta_{mp} = 1 \quad \text{and} \quad \Theta_{mp}' = \frac{\sigma_2 - \sigma_1}{\sigma_1} R_2 \Theta_s' \quad (33a)
\]

\[
\eta_{mp} = \eta_p; \quad \Theta_{mp} = \Theta_p \quad \text{and} \quad \Theta_{mp}' = \Theta_{md}' \quad (33b)
\]

(iv) Two-Phase Dispersed Region

\[
f_{md}^+ + \frac{(\sigma_2 - \sigma_1)^2}{2 Pr} \left( \frac{1 - F}{F_p} \right)^b f_{md} f_{md}^+ + \left( \eta_p - 1 + \frac{\sigma_1}{\sigma_2 - \sigma_1} \right) f_{md}'
\]

\[
\frac{b}{F_p(2 - \Theta_{mp} + k(\Theta_{mp} - 1))^2} \left( 1 - \frac{F}{F_p} \right) \frac{1}{\Theta_{md} f_{md}} = 0 \quad (34)
\]
reaches the packing limit fraction, the packing limit isotherm is controlled by these seven dimensionless parameters.

Evidently, the freeze-coating process under consideration is controlled by these seven dimensionless parameters appearing in the above equilibrium partition ratio

\[ Q \]

the molten substance superheating parameter

\[ Q \]

Supplemental Equation

\[ 5 \]

\[ 1 \]

(38)

(v) Molten Substance Region

\[ f''_m + \frac{\sigma_2^2}{2 \tau_f} f'_1 f'_1 = 0 \] (37)

\[ \Theta''_m + \frac{\sigma_2^2}{2 \tau_f} f'_1 \Theta'_m = 0 \] (38)

\[ \eta_m = \frac{\sigma_2 - \sigma_1}{\sigma_2} f_{md} - \left( 1 - \eta_p \right) \frac{\sigma_1}{\sigma_2 - \sigma_1} f_{md} \]

\[ f'_1 = f''_{md} \] (39a)

\[ f''_m = \frac{\sigma_2^2}{\sigma_2 - \sigma_1} f_{md}' , \quad \Theta'_m = \frac{\sigma_2}{\sigma_2 - \sigma_1} R_3 \Theta'_m \]

\[ \eta_m \to \infty: f'_1 = 0 \quad \text{and} \quad \Theta'_m = 3 \] (39b)

(vi) Supplemental Equation

\[ F = \frac{2 - \Theta_{md}}{(2 - \Theta_{md}) + \kappa (\Theta_{md} - 1)} \] (40a)

The supplemental equation can be used to determine the packing limit isotherm, \( \Theta_m \). At the location where the solid fraction reaches the packing limit fraction, the packing limit isotherm is

\[ \Theta_m = 2 - 2 F_p + \kappa F_p \] (40b)

There are seven dimensionless parameters appearing in the above system of similarity equations. These parameters are the freeze-coat-to-wall thermal ratio \( R_1 \), the wall subcooling parameter \( R_2 \), the molten substance superheating parameter \( R_3 \), the Prandtl number of the molten substance \( Pr \), the Stefan number \( Ste \), the equilibrium partition ratio \( \kappa \), and the exponent \( b \) in Eq. (34) that is directly proportional to the packing limit fraction \( F_p \):

\[ R_1 = \sqrt{\frac{k_p C_p}{k_{sw} C_{sw}}} , \quad R_2 = \frac{T_1 - T_p}{T_2 - T_1} , \quad R_3 = \frac{T_3 - T_2}{T_2 - T_1} \]

\[ Pr = \frac{v_l}{\alpha_l} , \quad Ste = \frac{C_p (T_2 - T_1)}{\Delta H_m} , \quad \kappa = \frac{m_2}{m_1} \quad \text{and} \quad b = [\mu] F_p = 2.5 F_p \]

Evidently, the freeze-coating process under consideration is controlled by these seven dimensionless parameters.

Solution Methodology

A closed-form solution for \( \Theta_w \) and \( \Theta_s \) can be obtained from Eqs. (28) and (30) by direct integration using the appropriate boundary conditions (Eqs. (29a–29b) and (31a–31b) respectively). This gives

\[ \Theta_w = \frac{R_1}{R_1 + \text{erf}(\sigma_1/2)} \] (42)

\[ \Theta_s = \frac{R_1}{R_1 + \text{erf}(\sigma_1/2)} \] (43)

The dimensionless temperature at the interface between the wall and the freeze-coating region, i.e., at \( \eta_w = \eta_s = 0 \), is given by

\[ \Theta_w(0) = \Theta_s(0) = \frac{R_1}{R_1 + \text{erf}(\sigma_1/2)} \] (44)

The first derivative of \( \Theta_s \) evaluating at \( \eta_s = 1 \) can be determined from Eq. (43) as

\[ \Theta'_s(1) = \frac{\sigma_1}{\sqrt{\pi}} \frac{\exp(-\sigma_1^2/4)}{R_1 + \text{erf}(\sigma_1/2)} \] (45)

Applying the above equation to the flux condition (Eq. (31b)) yields

\[ \Theta'_m(1) = \frac{(\sigma_2 - \sigma_1) \sigma_2}{\sqrt{\pi}} \frac{\exp(-\sigma_1^2/4)}{R_1 + \text{erf}(\sigma_1/2)} \] (46)

For a given set of parameters, equations (32), (34), (35), (37), and (38) with the appropriate boundary conditions can be numerically solved by using the classical fourth-order Runge-Kutta method. To avoid the singularity point at \( \eta_{md} = \eta_p \), the Taylor’s series expansion is applied to the momentum equation for the two-phase dispersed region, Eq. (34). In so doing, the initial condition is set at \( \eta_s = \Delta \eta_{md} \) instead of \( \eta_s \), where \( \Delta \eta_{md} \) is a small increment of \( \eta_{md} \). In order to solve the remaining equations, the values of \( \sigma_1 \), \( \sigma_2 \), and \( f''_{md}(\eta_s + \Delta \eta_{md}) \) are first estimated. Thereafter, the value of \( \eta_p \) can be obtained by numerically solving the heat conduction equation in the two-phase packing region using Eq. (46) as an initial condition. Once \( \eta_s \) is known, the momentum and energy equations for the two-phase dispersed and molten substance regions can be solved. The Secant iterative procedure is applied until the estimated values of \( \sigma_1 \), \( \sigma_2 \), and \( f''_{md}(\eta_s + \Delta \eta_{md}) \) match the boundary conditions given by Eqs. (36b) and (39b), i.e., \( \Theta_{md}(2) = 2 \), \( \Theta_s(\infty) = 3 \) and \( f'_s(\infty) = 0 \). The solution is shown to converge under the prescribed tolerance of \( 10^{-3} \).

Grid independency is also examined by setting the standard grid size at \( \Delta \eta_{mp} = \Delta \eta_{md} = 0.01 \) and \( \Delta \eta = 0.02 \) where the reference case \((R_1 = 1, R_2 = 10, R_3 = 1, Pr = 1, Ste = 0.1, \kappa = 0.3, F_p = 0.6, b = 1.5) \) is chosen to test the algorithm. With the grid size being increased by two and four times, the relative errors of the calculated values of \( \sigma_1 \) and \( \sigma_2 \) are less than 0.1 percent and 0.3 percent, respectively. On the other hand, as the grid size is decreased to one half of the standard grid size, the relative error is found to be less than 0.03 percent. This numerical accuracy is considered satisfactory for predicting the behavior of the freeze-coating process.

Results and Discussion

Numerical calculations have been made to determine the dependence of the solidus, packing limit and liquidus constants (\( \sigma_1 \), \( \sigma_p \), and \( \sigma_2 \)) on the controlling parameters. These three solidification constants characterize the behavior of the two-phase mushy zone. In practice, the wall subcooling parameter and the molten substance superheating parameter can be controlled by adjusting the inlet wall temperature and the ambient bath temperature, re-
spectively. On the other hand, the freeze-coat-to-wall thermal ratio, the Prandtl number, the Stefan number, the equilibrium partition ratio, and the packing limit fraction depend upon selection of the binary substance and wall material.

In this study, selected cases with various sets of values of the controlling parameters have been studied. Typical numerical results are depicted in Figs. 4 to 8. These results are obtained by varying the controlling parameters from the reference case. As expected, the liquidus constant is always greater than the solidus constant as the liquidus temperature, $T_1$, is higher than the solidus temperature, $T_2$. On the other hand, the packing limit constant lies between the values of the solidus and liquidus constants.

Figure 4 depicts the variations of the solidus, packing limit and liquidus constants with the freeze-coat-to-wall thermal ratio $R_1$ for different Stefan numbers. For a given Stefan number, $s_1$, $s_p$, and $s_2$ decrease with increasing $R_1$. As the thermal conductivity and the heat capacity of the wall compared to those of the freeze coat become smaller, the cooling capacity of the wall is reduced, resulting in a slower freeze-coating process. Note that as the value of $R_1$ approaches zero, corresponding to an infinite heat capacity of the wall, $s_1$, $s_p$, and $s_2$ asymptotically approach to constant values. On the other hand, for a given $R_1$, increasing the Stefan number causes $s_1$, $s_p$, and $s_2$ to increase. Under the same cooling conditions, a thicker freeze coat is expected for a material with a larger Stefan number.

The variations of the solidus, packing limit and liquidus constants with the wall subcooling parameter for different freeze-coat-to-wall thermal ratios are illustrated in Fig. 5. For a given thermal ratio, $s_1$, $s_p$, and $s_2$ increase with increasing $R_2$. Physically, an increase in $R_2$ corresponds to a higher value of the degree of subcooling, leading to a thicker freeze coat. On the other hand, for a given subcooling parameter, $s_1$, $s_p$, and $s_2$ increase as $R_1$ is decreased. For $R_1$ less than 0.01, the three solidification constants tend to approach their asymptotic values corresponding to those for an isothermal wall.

Figure 6 depicts the variation of the solidus, packing limit and liquidus constants with wall subcooling parameter for different Stefan numbers. In this figure, $R_1$ is set equal to zero, corresponding to an isothermal wall. For a given Stefan number, $s_1$, $s_p$, and $s_2$ increase with $R_2$, similar to the behavior shown in Fig. 5. Note that the values of $s_1$, $s_p$, and $s_2$ for an isothermal wall with infinite heat capacity represent the upper limits for the three solidification constants. For a given $R_2$, $s_1$, $s_p$, and $s_2$ increase with increasing Stefan number. Comparison of Figs. 4 and 6 indicates that $s_1$, $s_p$, and $s_2$ are more sensitive to the change in the values of $R_2$ than that of $R_1$.

The variations of the solidus, packing limit and liquidus constants with the molten substance superheating parameters for different Prandtl numbers are illustrated in Fig. 7. For a given Prandtl number, $s_1$, $s_p$, and $s_2$ decrease with increasing $R_3$. This is expected as more heat is convected from the liquid as the superheating parameter is increased, resulting in a slower freeze-coating process. Note that both $s_1$ and $s_p$ vary with $R_3$ in a manner which are quite different than that of $s_2$. As $R_3$ becomes smaller, both $s_1$ and $s_p$ asymptotically approach to constant values whereas $s_2$ continues to increase. For a given $R_3$, $s_1$, $s_p$, and $s_2$ increase as the Prandtl number is decreased. It is known that the convective heat transfer coefficient decreases with a decrease in the Prandtl number. Thus at a smaller value of the Prandtl number, the rate of heat convected from the liquid is reduced, resulting in a thicker freeze coat layer. Note that for Pr $> 10$, there is virtually no effect of the Prandtl number on $s_1$, $s_p$, and $s_2$.

Figure 8 depicts the variation of the solidus, packing limit and liquidus constants with the molten substance superheating parameter for different Stefan numbers for the case of a large Prandtl number fluid, i.e., Pr = 100. For a given Stefan number, $s_1$, $s_p$, and $s_2$ vary with $R_1$ in the same manner as those shown in Fig. 7. The asymptotic behavior of $s_1$ and $s_p$ as $R_1$ approaches zero is
observed for all Stefan numbers, but the same behavior is not observed for $\sigma_2$. Note that for Stefan number smaller than 0.02, $\sigma_1$ and $\sigma_p$ appear to be quite insensitive to the change of $R_3$. This trend, however, is not true for the liquidus constant $\sigma_2$. Comparison of Figs. 6 and 8 shows that $\sigma_1$, $\sigma_p$, and $\sigma_2$ are more sensitive to the change in the values of $R_2$ than that of $R_3$. The effects of the equilibrium partition ratio $\kappa$ and the packing limit fraction $F_p$ are presented in Tables 1 and 2, respectively. As the value of $\kappa$ is increased from 0.1 to 0.5, $\sigma_1$ increases by less than 4 percent whereas $\sigma_2$ increases by almost 15 percent. On the
other hand, $\sigma_p$ decreases by nearly 9 percent. Note from Eq. 40b that for a given $F_p$, the packing limit isotherm, $\theta_p$, decreases as $\kappa$ is increased, thus resulting in a smaller value of $\sigma_p$. The solidus and liquidus constants are weak functions of the packing limit fraction, as shown in Table 2. As the value of $F_p$ is increased from 0.5 to 0.75, $\sigma_1$ and $\sigma_2$ increase by less than 0.1 percent. In contrast, the value of $\sigma_p$ decreases by almost 12 percent over the same range of $F_p$. The sensitivity of $\sigma_p$ on $F_p$ is due to the fact that the packing limit isotherm depends directly on the packing limit fraction, as given by Eq. (40b).

Fig. 7 Variations of the solidus, packing limit and liquidus constants with the molten substance superheating parameter and the Prandtl number ($R_1 = 1, R_2 = 10, \text{Ste} = 0.1, \kappa = 0.3$, and $F_p = 0.6$)

Fig. 8 Variations of the solidus, packing limit and liquidus constants with the molten substance superheating parameter and the Stefan number for the high Prandtl number case ($R_1 = 1, R_2 = 10, \text{Pr} = 100, \kappa = 0.3$, and $F_p = 0.6$)
perheating parameter. the freeze-coat-to-wall thermal ratio and the molten substance superheating parameter, or the Prandtl number. For small Stefan numbers of the problem, are functions of seven independent of the Prandtl number. These three solidification constants, which represent the major unknowns of the problem, appear to be insensitive to the variation of the packing limit fraction.

Table 1 The Solidus, packing limit and liquidus constants for various values of the equilibrium partition ratio \( R_1 = 1, R_2 = 10, R_3 = 1, Pr = 1, Ste = 0.1, \) and \( F_p = 0.6 \)

<table>
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<tr>
<th>( k )</th>
<th>( \alpha_1 )</th>
<th>( \alpha_2 )</th>
<th>( \alpha_3 )</th>
<th>( \alpha_4 )</th>
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<td>0.6386</td>
<td>0.8097</td>
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</tr>
<tr>
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<td>0.8339</td>
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</tr>
<tr>
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<td>0.4606</td>
<td>0.6151</td>
<td>0.8547</td>
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</tr>
</tbody>
</table>

Table 2 The solidus, packing limit and liquidus constants for various values of the packing limit fraction \( R_1 = 1, R_2 = 10, R_3 = 1, Pr = 1, Ste = 0.1, \) and \( k = 0.3 \)

<table>
<thead>
<tr>
<th>( F_p )</th>
<th>( \alpha_1 )</th>
<th>( \alpha_2 )</th>
<th>( \alpha_3 )</th>
<th>( \alpha_4 )</th>
</tr>
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<td>0.6670</td>
<td>0.8096</td>
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<tr>
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</table>

Conclusions

A theoretical study of the process of freeze coating of a binary substance on a chilled moving plate has been performed. Based on the results of this study, the following conclusions can be drawn:

1. The behavior of the two-phase mushy zone can be characterized by the solidus constant, the packing limit constant, and the liquidus constant. These three solidification constants, which represent the major unknowns of the problem, are functions of seven dimensionless parameters of the freeze-coating system. These parameters are the freeze-coat-to-wall thermal ratio, the wall subcooling parameter, the molten substance superheating parameter, the Prandtl number, the Stefan number, the equilibrium partition ratio, and the packing limit fraction.

2. As the Stefan number or the wall subcooling parameter is increased, the solidus constant, the packing limit constant, and the liquidus constant increase. On the other hand, the three solidification constants decrease as either the freeze-coat-to-wall thermal ratio, the molten substance superheating parameter, or the Prandtl number is increased. At a sufficiently large Prandtl number (i.e., \( Pr > 10 \)), however, the three solidification constants appear to be independent of the Prandtl number. For small Stefan numbers (Ste < 0.02), the solidus constant and the packing limit constant are not sensitive to the variation of the molten substance superheating parameter.

3. For given values of the Prandtl number and Stefan number, the three solidification constants appear to be more sensitive to the variation in the wall subcooling parameter than to the variations in the freeze-coat-to-wall thermal ratio and the molten substance superheating parameter.

4. The three solidification constants approach asymptotically to constant values as the freeze-coat-to-wall thermal ratio \( R_1 \) approaches zero, corresponding to the case of an isothermal plate. Similarly, as the molten substance superheating parameter \( R_1 \) approaches zero, corresponding to the case of a saturated liquid, the solidus constant and the packing limit constant approach asymptotically to constant values. However, the liquidus constant continues to increase with decreasing value of the superheating parameter. Note that the maximum rate of freeze coating is obtained for the case of an isothermal plate (\( R_1 = 0 \)) moving through a saturated binary substance (\( R_1 = 0 \)).

5. Both the value of the packing limit constant and the liquidus constant change appreciably as the equilibrium partition ratio is varied from 0.1 to 0.5, which covers the range anticipated for most binary substances. On the other hand, the value of the solidus constant varies only slightly over this range. The packing limit constant tends to decrease as the packing limit fraction is increased. On the contrary, over the range considered in this study, both the solidus constant and liquidus constant appear to be insensitive to the variation of the packing limit fraction.

References


