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Technical Note

A similarity solution for solidification of an under-cooled binary alloy

V.R. Voller *

Saint Anthony Falls Laboratory, University of Minnesota, Civil Engineering, 500 Pillsbury Drive SE, Minneapolis, MN 55455, United States

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Abstract

A similarity solution for solidification of an under-cooled binary alloy melt in a semi-infinite, one-dimensional, insulated slot is developed. It is shown that this solution is a generalization of the known similarity solution for the solidification of an under-cooled pure melt, and is a special case of the similarity solution for solidification of a binary alloy previously presented in the literature. The new solution is used to quantify the effect of the Lewis number (ratio of thermal to solutal diffusivity) on the behavior of solidification. Limits on the amount of under-cooling that will admit a physically meaningful solution are obtained.

1. Introduction

There is significant current interest in modeling dendritic growth, see examples in [1–9]. The basic problem involves the free dendritic growth from a circular solid seed placed in a two-dimensional cavity containing an under-cooled melt of a pure material. A more advanced problem is to consider free dendritic growth into an under-cooled binary alloy [7–9], where, in addition to heat transfer, the transport of solute component needs to be considered. This latter problem is a true test of the state of the art of both solidification models and computational tools for phase change problems. In developing these computational solutions it is important to have appropriate analytical solutions so that rigorous verification can be carried out, limit behaviors investigated, and physical constraints identified.

In considering solidification problems which involve under-cooling and solute transport there are two known analytical similarity solutions in the literature. The first, due to Rubinstein [10] and investigated by Alexiades and Solomon [11], considers the one-dimensional solidification of a binary alloy. This problem is driven by the prescription of a fixed temperature at the origin $x = 0$, and in typical presentations in the literature [12, p. 248, 11, p. 106, 9] the initial condition is a melt at a uniform composition with a temperature at or above the liquidus, i.e., the melt is not under-cooled. The second problem, presented in Carslaw and Jaeger [13], is for the solidification of an under-cooled melt in a one-dimensional insulated semi-infinite slot. Initially the slot contains an under-cooled melt and solidification is initiated by introducing a solid seed at $x = 0$. In this second problem the melt is of a pure material, i.e., solute transport plays no role.

The objective of this work is to combine the concepts in the Rubinstein [10] and Carslaw and Jaeger [13] solutions to arrive at a similarity solution for the solidification of an under-cooled binary alloy, i.e., develop an analytical solution for solidification initiated by the placement of a solid seed crystal that includes both solute transport and under-cooling. Analysis is presented that shows that this new solution can be viewed as a generalization of the Carslaw and Jaeger [13] solution for the solidification of an under-cooled melt, and a special case of the Rubinstein [10] binary alloy similarity solution. In addition, the solution is used to investigate the influence of the Lewis number (ratio of thermal diffusivity to solutal diffusivity) on the

* Tel.: +1 612 625 0764; fax: +1 612 6267 7750.
E-mail address: volle001@umn.edu

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Nomenclature

C	solute concentration (wt%)
C_0	initial solute concentration (wt%)
c	specific heat (J/kg K)
D	mass diffusivity (m^2/s)
ℓ	length scale (m)
L	latent heat (J/kg)
$Le = \alpha_l/D_l$	Lewis number
m	slope of the liquidus line in the binary phase diagram
$M = -mc/L$	dimensionless liquidus slope
s	position of solid–liquid interface
T	temperature (K)
T_f	fusion temperature of the solvent

Greek symbols

α	thermal diffusivity in the liquid phase (m^2/s)
λ	similarity variable

Superscript

*	dimensioned quantity

Subscripts

s	solid phase value
l	liquid phase value

solidification behavior and to identify physical limit conditions for the problem.

2. Problem and governing equations

The problem domain consists of a one-dimensional semi-infinite slot. Initially the slot is filled with an alloy melt with a uniform composition and temperature below the liquidus temperature of the alloy i.e., the slot contains an under-cooled alloy melt. Solidification is initiated by placing a thin solid layer, at the solidification equilibrium temperature, at $x = 0$. If curvature (the slot domain is narrow) and kinetic effects are neglected, and there is sufficient latent heat to over-balance the sensible heat in the melt, the temperature of the liquid layer immediately adjacent to the solid seed layer will increase to come into equilibrium with the solid. This results in a negative temperature gradient out of the liquid layer, which drives additional solidification by removing the residual latent heat. In this way, the solid layer will grow into the slot. In a binary alloy, as new solid forms solute is rejected into the liquid phase. This addition of solute into the liquid ahead of the solidification front will control (reduce) the equilibrium temperature at which solid forms.

To arrive at governing equations for the above problem the following dimensionless numbers are introduced:

$$T = \frac{T^* - T_f - mC_0}{L/c_l}, \quad C = \frac{C^*}{C_0}, \quad x = \frac{x^*}{\ell}, \quad s = \frac{s^*}{\ell}, \quad t = \frac{\alpha_l t^*}{\ell^2} \tag{1}$$

where the superscript (*) indicates a dimensioned quantity, T^* (K) is temperature, T_f is the fusion temperature of the solvent, $m < 0$ is the slope of the liquidus line in the binary phase diagram, C^* (wt%) is the solute concentration, C_0 is the initial solute concentration in the melt, L (J/kg) is the latent heat, c_l (J/kg K) is the specific heat in the liquid phase, s^* (m) is the position of the sharp interface between the solid and the liquid α_l (m^2/s) is the thermal diffusivity in the liquid phase, and ℓ (m) is a convenient length scale. With these definitions the governing equations are:

Heat transport in the solid:

$$\frac{\partial T}{\partial t} = \frac{\alpha_s}{\alpha_l}\frac{\partial^2 T}{\partial x^2}, \quad 0 \leqslant x \leqslant s(t) \tag{2}$$

Heat transport in the liquid:

$$\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2}, \quad x \geqslant s(t) \tag{3}$$

Solute transport in the solid:

$$\frac{\partial C_s}{\partial t} = \frac{1}{Le}\frac{D_s}{D_l}\frac{\partial^2 C_s}{\partial x^2}, \quad 0 \leqslant x \leqslant s(t) \tag{4}$$

Solute transport in the liquid:

$$\frac{\partial C_l}{\partial t} = \frac{1}{Le}\frac{\partial^2 C_l}{\partial x^2}, \quad x \geqslant s(t) \tag{5}$$

where the subscript s refers to the solid phase, the subscript l to the liquid phase, D is the mass diffusivity (m^2/s) and $Le = \alpha_l/D_l$ is the Lewis number.

Boundary conditions are: at $x = 0$

$$\frac{\partial C_s}{\partial x} = 0, \quad \frac{\partial T}{\partial x} = 0 \tag{6}$$

as $x \to \infty$

$$C_l \to 1, \quad T \to T_0 < 0 \tag{7}$$

where $T_0 < 0$ is the initial under-cooled temperature.

On the moving solid/liquid interface at $x = s(t)$

$$C_l = C_i(t), \quad C_s = kC_i(t), \quad T = T_i(t) = MC_0(1 - C_i) \tag{8}$$

where k is the partition coefficient (assumed constant) and $M = -mc_l/L$,

$$\frac{\alpha_s}{\alpha_l}\frac{\partial T}{\partial x}\bigg|_{s^-} - \frac{\partial T}{\partial x}\bigg|_{s^+} = \frac{ds}{dt} \tag{9}$$

$$\frac{D_s}{D_l}\frac{1}{Le}\frac{\partial C_s}{\partial x} - \frac{1}{Le}\frac{\partial C}{\partial x} = (1-k)C_i\frac{ds}{dt} \tag{10}$$

the last two conditions expressing the heat (Stefan condition) and mass balance at the interface.

3. A similarity solution

A similarity solution is sought in which the interface movement is given by

$$s = 2\lambda\sqrt{t} \tag{11}$$

and the interface concentration C_i and temperature T_i are constants. Due to the fact that the slot is insulated this last condition immediately leads to the solid phase solutions $T = T_i$ and $C_s = C_i$. The temperature and concentration solutions in the liquid phase are

$$T = T_0 + (T_i - T_0)\frac{\operatorname{erfc}\left(\frac{x}{2\sqrt{t}}\right)}{\operatorname{erfc}(\lambda)} \tag{12}$$

and

$$C = 1 + (C_i - 1)\frac{\operatorname{erfc}\left(\frac{x\sqrt{Le}}{2\sqrt{t}}\right)}{\operatorname{erfc}\left(\lambda\sqrt{Le}\right)} \tag{13}$$

The unknown values C_i, T_i and λ in (11)–(13), are found from the simultaneous solution of the equation of the liquidus line (8), the interface heat balance (9), and the interface mass balance (10), i.e.,

$$T_i - MC_0(1 - C_i) = 0 \tag{14}$$

$$\lambda\sqrt{\pi}e^{\lambda^2}\operatorname{erfc}(\lambda) - (T_i - T_0) = 0 \tag{15}$$

$$(1-k)C_i\lambda\sqrt{Le}\sqrt{\pi}e^{\lambda^2 Le}\operatorname{erfc}(\lambda\sqrt{Le}) - (C_i - 1) = 0 \tag{16}$$

4. Analysis

4.1. Relationship to previous solutions

The development of the solution in (11)–(16) closely follows the approaches used in the solutions by Carslaw and Jaeger [13] for solidification of a pure under-cooled melt, and by Rubinstein [10] for the solidification of a binary alloy. In this context it is worthwhile to establish explicit connections between the solution (11)–(16) and those previously presented in the literature.

In the first place it is noted that a setting of $k = 1$ eliminates solute segregation and solute gradients. With this setting (13) and (16) set a constant concentration $C = C_i = 1$, throughout the domain and (14) establishes an interface temperature of $T_i = 0$. The net result, is a reduction of (11)–(16) to the three equations

$$s = 2\lambda\sqrt{t}, \quad T = T_0 - T_0\frac{\operatorname{erfc}\left(\frac{x}{2\sqrt{t}}\right)}{\operatorname{erfc}(\lambda)}, \quad (x > s(t)),$$

$$\lambda\sqrt{\pi}e^{\lambda^2}\operatorname{erfc}(\lambda) + T_0 = 0 \tag{17}$$

that exactly match those derived in the solution by Carslaw and Jaeger [13] for solidification of a pure under-cooled melt. Hence the solution (11)–(16) can be viewed as a generalization of the Carslaw and Jaeger [13] solution.

On the other hand, although the Rubinstein [10] solution is usually presented with an initial condition that explicitly sets the liquid temperature at or above the equilibrium liquidus temperature [9,11,12], thereby ruling out under-cooling, there is no reason why the solution cannot account for under-cooling. The governing transport equations for the Rubinstein problem with under-cooling match those used in this work, i.e., (2)–(5). The only difference in the problem formulation is the thermal condition at $x = 0$. In the Rubinstein problem the insulated condition $\partial T/\partial x = 0$ in (6) needs to be replaced by the Dirichlet condition $T(0,t) = T_{sur} \leqslant T_i$. Satisfying this alternative boundary condition modifies the solution (11)–(16) by adding the thermal profile

$$T_{sur} + (T_i - T_{sur})\frac{\operatorname{erf}\left(\frac{\sqrt{\alpha_l}}{\sqrt{\alpha_s}}\frac{x}{2\sqrt{t}}\right)}{\operatorname{erf}\left(\frac{\lambda\sqrt{\alpha_l}}{\sqrt{\alpha_s}}\right)} \tag{18}$$

which results in the updated thermal balance

$$\sqrt{\pi}\lambda - \sqrt{\frac{\alpha_s}{\alpha_l}}\frac{T_i - T_{sur}}{\operatorname{erf}\left(\frac{\lambda\sqrt{\alpha_l}}{\sqrt{\alpha_s}}\right)}e^{-\frac{\lambda^2\alpha_l}{\alpha_s}} - \frac{T_i - T_0}{\operatorname{erf}(\lambda)}e^{-\lambda^2} = 0 \tag{19}$$

in place of (15). The original solution (11)–(16), satisfying the insulated condition at $x = 0$ can be readily recovered, however, on simply setting $T_{sur} = T_i$ in (18) and (19). Hence the proposed solution (11)–(16) can be viewed as a special case of the Rubinstein solution which has been extended to account for under-cooling.

4.2. Effect of Lewis number

The most relevant parameter in the solution (11)–(16) is the Lewis number Le. This dimensionless parameter characterizes the relative roles of the thermal and solute diffusion in controlling the solid–liquid interface movement. The first panel in Fig. 1 shows the solute and temperature profiles at dimensionless time $t = 100$ for a case where $Le = 1$. As might be expected, the thermal and solutal boundary layers have similar thicknesses. If the Lewis number is decreased (see the second panel in Fig. 1 where $Le = 0.1$) however, the rate of the solute diffusion away from the interface increases, the "pile up" of solute at the solid–liquid interface is reduced, and the solutal boundary layer becomes thicker than the thermal boundary layer. On the other hand, if the Lewis number is increased (see the third panel in Fig. 1 where $Le = 10$) the rate of solute diffusion is reduced, the interface solute pile up increases and the solutal boundary layer is thinner than the thermal boundary layer.

To fully understand the limit behaviors at large and small Lewis numbers consider the normalized variables T_i/T_0 and λ/λ_0, where through $s_0 = \lambda_0\sqrt{t}$ the parameter λ_0 determines the solidification front in a pure melt initially under-cooled to T_0. For a given set of conditions, Fig. 2

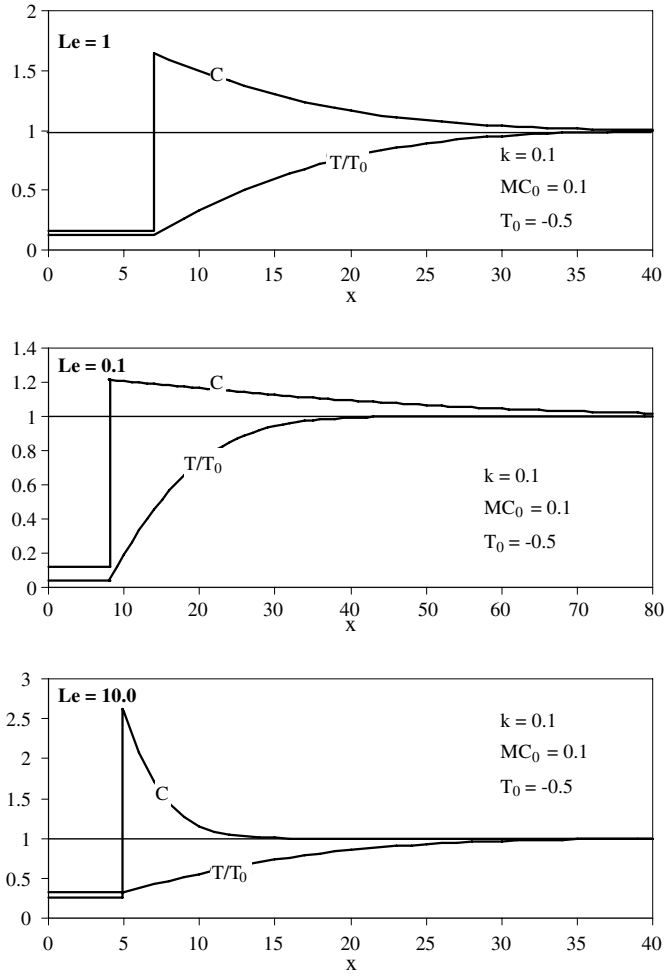

Fig. 1. Concentration and temperature profiles at time $t = 100$ for various Lewis numbers.

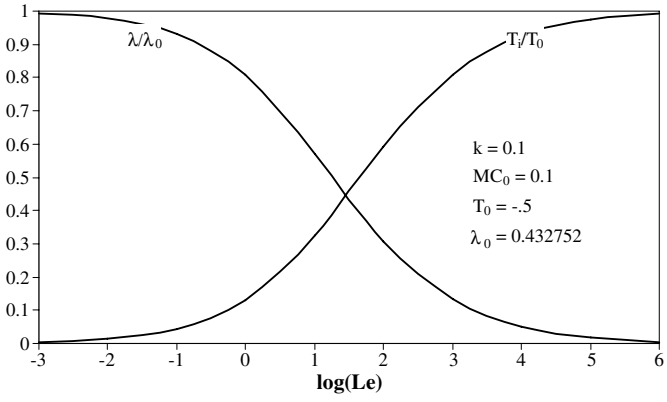

Fig. 2. Influence of Lewis number on interface movement and temperature.

plots these normalized variables across nine decades of Lewis number. As noted above, for small Lewis numbers the diffusion of solute away from the interface is rapid. In the limit of $Le \to 0$ the diffusion is infinitely fast and the solute in the liquid adjacent to the solid–liquid interface remains at the initial concentration, i.e., $C_i = C_0$ and $T_i = 0$. In this case the solution (11)–(16) reduces to the solution for an under-cooled pure material, see (17), previously presented in Carslaw and Jaeger [13]. At the opposite extreme, as $Le \to \infty$ the rate of solute diffusion away from the interface is very slow. This results in a slowing of the front movement $\lambda \to 0$ and, an increase in the interface temperature towards the maximum, i.e., $T_i \to T_0$.

4.3. Physical constraints on the solution

Extending an analysis presented by Alexiades and Solomon [11], it is noted that the solution (12)–(16) is only physically meaningful across a specified range of prescribed under-cooling values. Alexiades and Solomon [11] note that for physically meaningful values $\lambda > 0$, the first term in (15) is bounded by $0 \leqslant \lambda\sqrt{\pi}e^{\lambda^2}\mathrm{erfc}(\lambda) \leqslant 1$, hence $T_i \geqslant T_0 \geqslant T_i - 1$. On noting that, in the limit $\lambda \to 0$ (16) and (14) in turn give $C_i = 1$ and $T_i = 0$, whereas in the limit $\lambda \to \infty$ these equations give $C_i = 1/k$ and $T_i = MC_0 - MC_0/k$, the bounds on the under-cooling can be written as

$$0 \geqslant T_0 \geqslant MC_0 - \frac{MC_0}{k} - 1 \tag{20}$$

The physical interpretation is that, an under-cooling approaching the upper limit ($T_0 \to 0$) would lose the driving potential for the solidification, and an under-cooling approaching the lower limit would not have enough residual latent heat to bring the liquid layer, adjacent to the solid seed, to the equilibrium temperature. In the case of a pure material ($C_0 = 1$) or no solute partitioning ($k = 1$) the lower limit on the dimensionless under-cooling is -1. Eq. (20) shows that the introduction of a binary alloy with partitioning ($k < 1$) reduces this lower limit. In essence, with a binary alloy, since the solute rejected at the interface lowers the equilibrium temperature, less heat is required to bring the liquid layer to equilibrium and the magnitude of the under-cooling can be taken below the pure material limit of $T_0 = -1$.

5. Conclusions

This paper has presented a similarity solution for the solidification of an under-cooled binary alloy melt contained in a semi-infinite insulated slot. Solidification is initiated by introducing, at the origin, a solid seed at the equilibrium temperature. Previously presented and closely related solutions consider the solidification of a binary alloy under a prescribed temperature at the origin [10] or the solidification of an under-cooled pure material [13] from an initial solid seed. The combination of the binary alloy and under-cooling in one problem and solution provides a suitable test for the development of computational algorithms designed to simulate the solidification of under-cooled binary alloys. In addition, the proposed similarity solution provides important quantitative insights in to the relative roles of thermal and solute diffusive transport during solidification (see Figs. 1 and 2), and identifies

physical limits on the maximum amount of under-cooling that will allow for growth from a solid seed, see (20).

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