

# A boundary integral approach to the solidification of dilute alloys

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**Abstract**—We describe a boundary integral approach to solidification problems for which the Stefan number, and transition temperature, depend on time. The approach is applied to a simple model of alloy solidification and some sample problems are solved.

## 1. INTRODUCTION

THE SOLIDIFICATION of alloys is an extremely complex process that is difficult to model, even for situations involving only one spatial dimension [1, 2]. Besides the phenomena of heat conduction, phase transition and mass diffusion, the process can often involve recirculation in the melt region. Because of this complexity, some simplified models have been developed [3]. One such model, the 'well-stirred' model, attempts to take some account of both heat and mass transport. For the melt region, the model assumes the heat and mass transport to be so rapid as to cause the composition, and temperature, to be spatially uniform. However, within the solid, the mass transport is assumed negligible and the temperature taken to satisfy the full heat equation. Whilst by no means perfect, the model gives a reasonable representation of several solidification processes. Consideration of these ideas can be found in the book by Chalmers [3] and, more recently, in a paper by Alexiades [4].

The paper by Alexiades applies the well-stirred model to some problems in one dimension. However, many important processes cannot be simplified to this extent. Consequently, it is the purpose of this paper to develop a method for applying the well-stirred model to problems with two spatial dimensions. Under suitable conditions, the well-stirred model can be regarded as a Stefan problem with the Stefan number, and transition temperature, depending on time. In Section 2, it is shown how such problems can be solved using a boundary integral approach. The approach is an extension of the one described in ref. [5]. Section 3 considers the well-stirred model and Section 4 describes the solution of some sample problems.

## 2. THE BOUNDARY INTEGRAL EQUATIONS

Consider the problem of finding a temperature field  $T$  that satisfies the two-dimensional heat equation

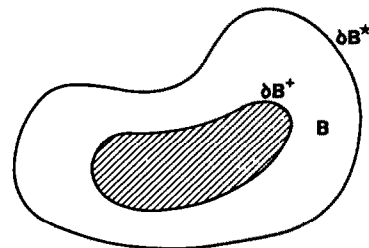


FIG. 1. Region  $B$ .

$$\left( \frac{\rho c}{k} \frac{\partial}{\partial t} - \frac{\partial^2}{\partial x^2} - \frac{\partial^2}{\partial y^2} \right) T = 0 \quad (1)$$

over a region  $B$  that is bounded by a fixed curve  $\partial B^*$  and a moving curve  $\partial B^+$  (Fig. 1). It is assumed that, at each point of  $\partial B^*$ , the temperature or heat flux is specified. Further, it is assumed that  $\partial B^+$  constitutes a phase change on which the conditions

$$T = T_i(t) \quad (2)$$

and

$$\frac{\partial T}{\partial n} = \alpha(t)v_n \quad (3)$$

are satisfied. Quantity  $\partial T / \partial n$  is the normal derivative of temperature on  $\partial B^+$  and  $v_n$  is the normal velocity of  $\partial B^+$ . The above problem is of the Stefan type, but with the Stefan number, and transition temperature, depending on time.

Space-time scales are chosen for which  $\rho c / k$  is unity. Further, the moving boundary is interpreted as a surface  $S^+$  in space-time and assumed to satisfy an equation of the form

$$t - f(x, y) = 0. \quad (4)$$

Boundary  $\partial B^*$  may be regarded as a cylindrical surface  $S^*$  in space-time and is taken to coincide with  $\partial B^+$  at time  $t = 0$ .

Let  $G$  be the function

$$G(\mathbf{r}, \mathbf{r}', t, t') = H(t - t') \exp \left[ \frac{-|\mathbf{r} - \mathbf{r}'|^2}{4(t - t')} \right] / [4\pi(t - t')]$$



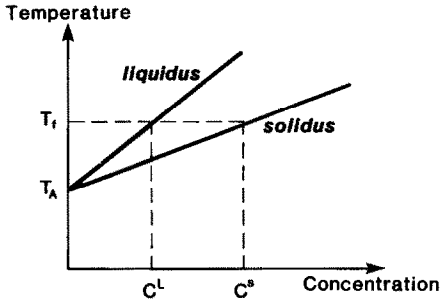


FIG. 2. The phase diagram.

with  $P^+$  at time  $t = 0$ .) Time is divided into intervals of duration  $\Delta t$  and, during any interval, each side of  $P^*$  will trace out a space-time quadrilateral. On such quadrilaterals,  $T$  and  $\partial T/\partial n$  are approximated by their centroidal values. Collocation of equation (8) at each quadrilateral centroid then produces a set of equations that determines the approximation.

The solution proceeds in a step-by-step fashion. At the  $i$ th step, the approximate boundary fields, and  $P^+$ , are assumed to be known for times from 0 to  $(i-1)\Delta t$ . The shape of  $P^+$  at time  $i\Delta t$  is estimated by extrapolation from the corresponding polygons at times  $(i-1)\Delta t$  and  $(i-2)\Delta t$ . Then, for the time interval from  $(i-1)\Delta t$  to  $i\Delta t$ , the approximate boundary fields are derived from a suitably discretized version of equation (8). (A suitable version of equation (8) will also provide the global temperature field.) An improved estimate for polygon  $P^+$  at time  $i\Delta t$  is found in the following fashion. For each vertex, a search is made of the line that joins the corresponding vertex at time  $(i-1)\Delta t$  to the mean position of these vertices. The improved vertex will then correspond to the point at which the phase-change temperature occurs. Once all the vertices have been improved, the boundary fields are recalculated and the vertices readjusted if further refinement is required. Obviously, for the first step, the extrapolation procedure is invalid. Consequently, in this case, the vertex positions are estimated by assuming each side to have moved according to the relevant planar solution [6]. (Further details of the numerical procedure may be found in ref. [5].)

### 3. DILUTE ALLOYS

Consider the two-dimensional solidification of a binary alloy. Assuming the concentration  $C$  (weight fraction) of one component (the impurity) to be weak, it is possible to approximate the phase diagram by that shown in Fig. 2. Let  $C^s$  and  $C^l$  denote the concentrations of impurity in the solid and liquid phases, respectively. When transition takes place

$$C^s = \frac{1}{\gamma_s} (T_f - T_A) \quad (9)$$

and

$$C^l = \frac{1}{\gamma_l} (T_f - T_A) \quad (10)$$

where  $\gamma_s$  and  $\gamma_l$  are the slopes of the straight lines.

The melt region is assumed to be 'well stirred', which means that a change at any point in the melt will be rapidly averaged out over the whole melt. Consequently,  $C$  and  $T$  will be uniform throughout the melt. Further, when solidification begins, the melt will have already cooled to its transition temperature. Once a given point has solidified, the concentration  $C$  is assumed to be frozen at its value for the instant of solidification (no movement of impurity in the solid phase).

Let  $A_0$  be the initial cross-sectional area of the melt and let  $A$  be the area at time  $t$ . In moving from solid to liquid phases, there is a jump in concentration. Consequently, if the melt area changes by  $dA$ , the concentration will change by

$$dC^l = [C] dA/A$$

where  $[C] = C^s - C^l$ . It is obvious that  $\gamma_s C^s = \gamma_l C^l$  and so

$$[C] = C^l \left( \frac{\gamma_l}{\gamma_s} - 1 \right).$$

Consequently

$$\frac{dC^l}{C^l} = \left( \frac{\gamma_l}{\gamma_s} - 1 \right) \frac{dA}{A}$$

which yields

$$C^l = \left( \frac{A}{A_0} \right)^\beta C_0^l \quad (11)$$

where  $\beta = \gamma_l/\gamma_s - 1$  and  $C_0^l$  is the initial concentration of impurity. Further, the transition temperature will be given by

$$T_f(t) = T_A + \gamma_l \left( \frac{A}{A_0} \right)^\beta C_0^l. \quad (12)$$

Consider the situation where a length  $ds$  of the solidification front advances a distance  $dn$ . There will be a change  $dn ds$  in melt area and so a temperature drop of

$$\gamma_l \beta \left( \frac{A}{A_0} \right)^\beta \frac{C_0^l}{A} dn ds$$

in the melt. For each unit length perpendicular to the plane, the heat liberated by this drop will be

$$c^l \rho^l \gamma_l \beta \left( \frac{A}{A_0} \right)^\beta C_0^l dn ds$$

where  $\rho^l$  is the density of the melt and  $c^l$  is its specific heat. However, because of the phase change, there will be an additional amount of heat liberated. If  $L$  is the latent heat of fusion, this additional heat will be given by  $\rho^l L dn ds$ . The liberated heat will be absorbed by the solid phase. Consequently, on the solid side of the phase change

$$k \frac{\partial T}{\partial n} = \rho^l L \frac{dn}{dt} + \gamma_1 c^l \rho^l \beta \left( \frac{A}{A_0} \right)^\beta C_0^l \frac{dn}{dt}$$

where  $k$  is the conductivity of the solid phase. Here we have assumed that, for a temperature drop due to impurity extraction, the heat is removed at the same site as the impurity.

In the notation of the previous section

$$\alpha(t) = \frac{\rho^l L}{k} + \gamma_1 \left( \frac{c^l \rho^l}{k} \right) \beta \left( \frac{A}{A_0} \right)^\beta C_0^l.$$

**4. SOME EXAMPLES**

Consider the solidification of a copper-nickel alloy [4]. For such an alloy,  $T_A = 1083^\circ\text{C}$ ,  $\gamma_1 = 640^\circ\text{C}$  and  $\gamma_s = 320^\circ\text{C}$ . During solidification, the body surface is assumed to lose heat at a rate proportional to  $T - T_E$  where  $T_E$  is the temperature of the external environment. For the current simulations,  $T_E$  is taken to be  $0^\circ\text{C}$ . At the instant before solidification starts, the melt is assumed to have a 10% concentration of nickel and so  $T_1$ , the melt temperature at the start of solidification, will be  $1147^\circ\text{C}$ . A new temperature scale is introduced for which  $T_1$  is zero and  $T_1 - T_E$  is unity. Then, in terms of this scale,  $\gamma_1$  will assume the value 0.55798 and  $\gamma_s$  the value 0.27899. Space-time scales are introduced for which  $\rho c/k$  becomes unity and

$\rho^l c^l/k$  is also assumed to be unity. Consequently, since  $L/c^l = 410^\circ\text{C}$  for the present alloy [4],  $\rho^l L/k$  will assume the value 0.35745 in terms of the new scales. The boundary condition at the body surface will be

$$\frac{\partial T}{\partial n} = K(T - T_E)$$

and  $K$  is chosen such that  $\partial T/\partial n|_{r=0} = -1$ .

Consider the solidification of an infinite rod of square cross-section, each side of the square having length 2. For discretization purposes, the boundary of the cross-section is divided into 80 equal length intervals and time into intervals of duration 0.04. Although the discretization would appear to introduce 80 unknowns at each time step (the 80 mid-segment values of  $T$ ), these are reduced to 10 by virtue of the symmetry in the problem. Simulations using the methods of Section 2 predict the phase change to move in the fashion shown in Fig. 3(a). The figure shows the top right-hand corner of the rods cross-section with phase-change curves sampled at times 0.0, 0.12, 0.24, 0.36 and 0.48. For the final solid, each curve will constitute a contour of constant concentration, the respective concentrations being 20, 12.16, 7.27, 3.76 and 1.36%. Figure 4(a) shows the temperature distribution on a half side of the cross-section.

To check the validity of the simulation, it was repeated with a time step of 0.02 and the boundary divided into 120 equal length intervals. The original

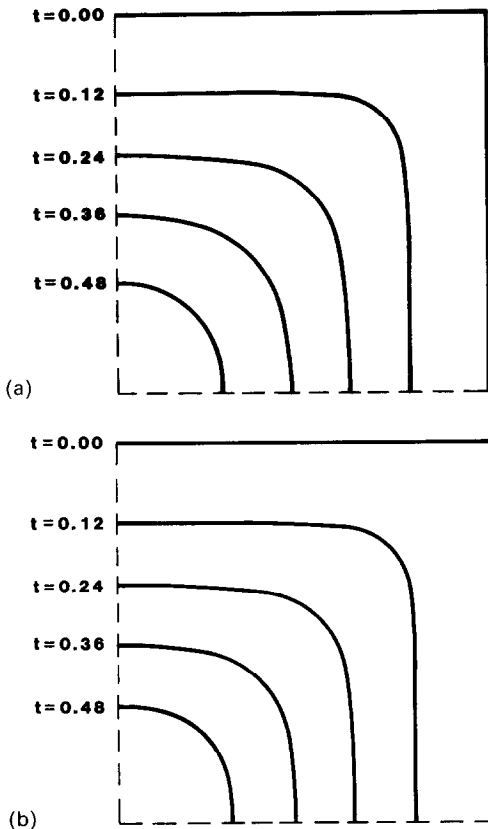


FIG. 3. The phase-change fronts for linear cooling: (a) the well-stirred melt; (b) fixed  $\alpha$  and  $T_f$ .

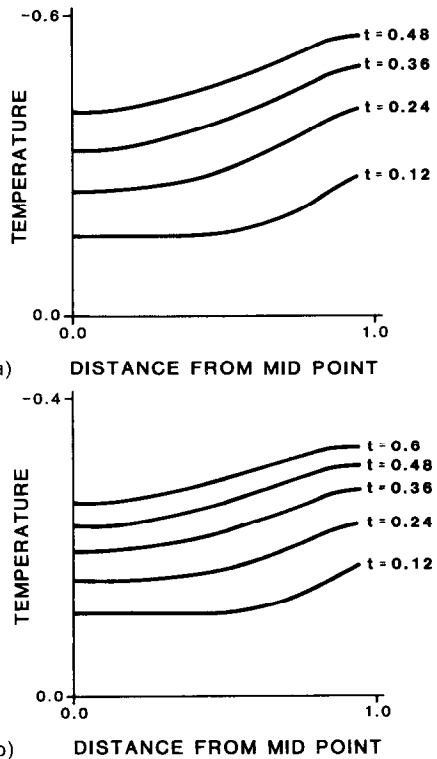


FIG. 4. The temperature distribution on the boundary: (a) linear cooling; (b) non-linear cooling.

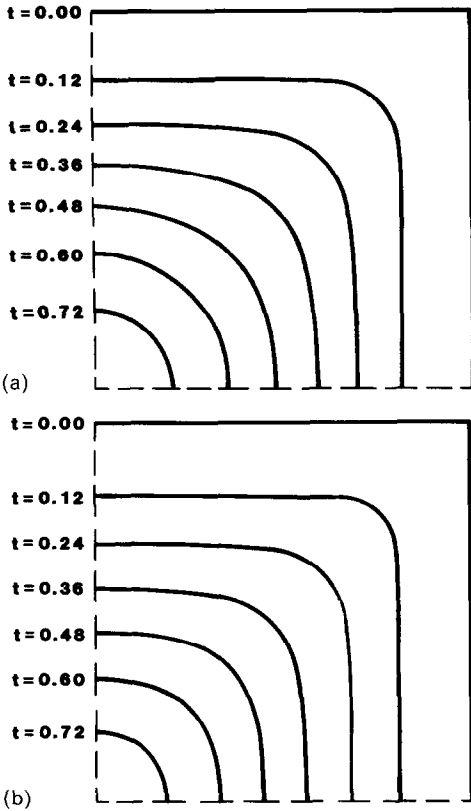


FIG. 5. The phase-change fronts for non-linear cooling: (a) the well-stirred melt; (b) fixed  $\alpha$  and  $T_f$ .

simulation predicted a time of 0.611 to complete solidification whilst the more refined discretization predicted a time of 0.602. Further, at time 0.48, the phase-change curves were within a distance 0.008 of each other.

To gauge the effect of the well-stirred model, the above simulations were repeated with constant  $\alpha(t)$  and  $T_f(t)$  (values being chosen to coincide with  $\alpha(0)$  and  $T_f(0)$  from the above simulation). The phase-change curves are shown in Fig. 3(b), the sample times being the same as for Fig. 3(a). It can be seen that the only effect of the well-stirred model is to cause a slightly earlier rounding of the melt pool. However, this effect is accentuated if the rod surface loses heat in the fashion of a black body radiator, that is

$$\frac{\partial T}{\partial n} = K(T^4 - T_E^4)$$

where  $K$  is a constant and temperatures are measured from absolute zero. Consider a simulation for which  $K$  is chosen such that  $\partial T/\partial n|_{t=0} = -1$ , temperatures rescaled as before. For a well-stirred melt, the movement of the phase change is illustrated in Fig. 5(a). The figure shows the top right-hand corner of the cross-section with phase-change curves sampled at times 0.0, 0.12, 0.24, 0.36, 0.48, 0.6 and 0.72. For the final solid, each curve will constitute a contour of constant concentration, the respective concentrations being 20, 13.13, 9.20, 6.19, 3.82, 1.99 and 0.67%.

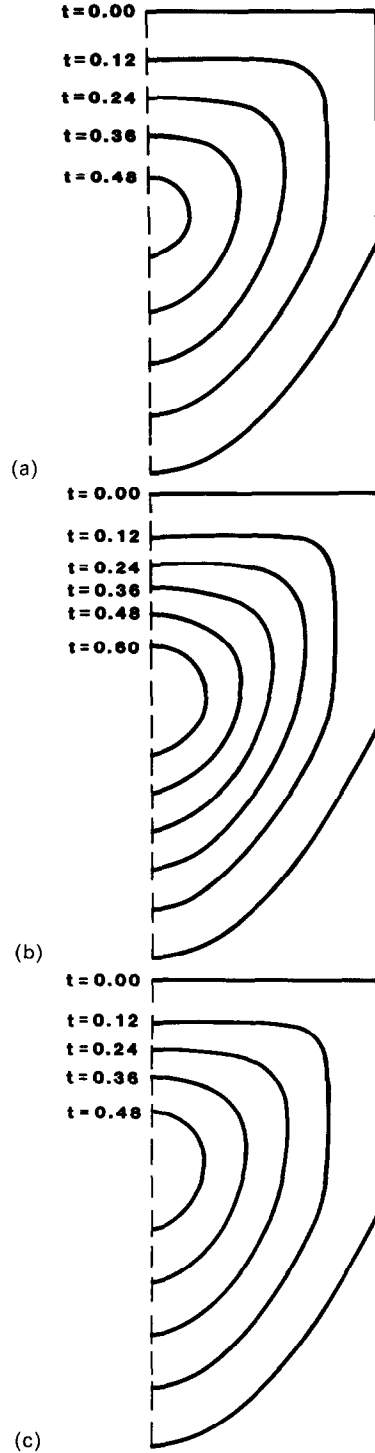


FIG. 6. The phase-change fronts for a well-stirred melt: (a) linear cooling; (b) non-linear cooling; (c) mixed cooling.

Figure 4(b) shows the temperature distribution on a half side of the cross-section and Fig. 5(b) shows the corresponding phase-change curves for a simulation with  $\alpha(t)$  and  $T_f(t)$  held constant.

As a further example, consider the solidification of a rod whose cross-section is bounded by the curves  $y = x^2$ ,  $x = -1$ ,  $x = 1$  and  $y = 2$ . For discretization

purposes, the linear parts of the boundary are divided into 40 equal length segments and the curved part into 32 equal length segments. Time is divided into intervals of duration 0.04. The melt is assumed to be well stirred and solidification is considered for three different sets of boundary conditions. Figures 6(a)–(c) show the simulated development of the phase-change curves for that part of the rod cross-section with  $x \geq 0$ . Figure 6(a) shows the development with boundary condition

$$\frac{\partial T}{\partial n} = K(T - T_E)$$

and  $K$  chosen such that  $\partial T/\partial n|_{t=0} = -1$ . Figure 6(b) shows the development with boundary condition

$$\frac{\partial T}{\partial n} = K(T^4 - T_E^4)$$

and  $K$  chosen such that  $\partial T/\partial n|_{t=0} = -1$ . Finally, Fig. 6(c) shows the development when the boundary at

$y = 2$  is subject to the above non-linear condition and the rest of the boundary subject to the linear condition. For Figs. 6(a) and (c), the sample times are 0.0, 0.12, 0.24, 0.36 and 0.48. Figure 6(b) includes an additional sample at time 0.6.

## REFERENCES

1. L. N. Tao, On solidification of a binary alloy, *Q. J. Mech. Appl. Math.* **33**, 211–225 (1980).
2. A. B. Crowley and J. R. Ockendon, On the numerical solution of an alloy solidification problem, *Int. J. Heat Mass Transfer* **22**, 941–948 (1979).
3. B. Chalmers, *Principles of Solidification*, 1st Edn, Chap. 5. Wiley, New York (1964).
4. V. Alexiades, Rapid freezing of dilute alloys, *IMA J. Appl. Math.* **30**, 67–79 (1983).
5. C. J. Coleman, A boundary integral formulation of the Stefan problem, *Appl. Math. Modelling* **10**, 445–449 (1986).
6. H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids*, 2nd Edn, Chap. 11. Oxford University Press, London (1973).

## UNE APPROCHE INTEGRALE LIMITE POUR LA SOLIDIFICATION D'ALLIAGES DILUES

**Résumé**—On décrit une approche intégrale limite pour les problèmes de solidification dans lesquels le nombre de Stefan et la température de transition dépendent du temps. L'approche est appliquée à un modèle simple de solidification d'alliage et à quelques problèmes types.

## EINE GRENZ-INTEGRAL-NÄHERUNG FÜR DIE ERSTARRUNG VON VERDÜNNTEN LEGIERUNGEN

**Zusammenfassung**—Wir beschreiben eine Grenzintegral-Näherung für Erstarrungsprobleme, bei welchen die Stefan-Zahl und die Übergangstemperatur von der Zeit abhängen. Die Näherung wird auf ein einfaches Modell der Erstarrung von Legierungen angewandt, einige Muster-Probleme werden gelöst.

## ПРИМЕНЕНИЕ ИНТЕГРАЛЬНОГО МЕТОДА ДЛЯ РЕШЕНИЯ ЗАДАЧ О ЗАТВЕРДЕВАНИИ СПЛАВОВ

**Аннотация**—Интегральный метод предлагается применять для решения задач затвердевания сплавов с зависящими от времени числом Стефана и температурой перехода. Метод проверяется на простой модели. Приведены примеры решения некоторых задач.