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CRYSTAL GROWTH WITH  
PARABOLIC INTERFACES



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Date Transmitted : August 1977

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## ABSTRACT

A mathematical treatment is presented for the growth of dendrites or precipitates with parabolic interfaces. Two cases are considered in which the interfaces correspond to a paraboloid of revolution and a parabolic cylinder. For these interfaces, relationships between growth rate and supercooling are obtained for two general boundary conditions under the assumption that the temperature or concentration variation along the interface is finite and continuous.

## 1. Introduction

Parabolic interfaces are very frequently encountered in situations involving crystal growth and solid state phase transformations. Important examples of these include dendrite growth, whisker or filament growth, and Widmanstätten plate and needle growth. The growth of precipitate particles during solid state transformation is primarily controlled by the diffusion of atoms, whereas the freezing or solidification process is governed by the conduction of heat as well as by the diffusion of atoms.

Considerable experimental work has been carried out to study the growth of dendrites and Widmanstätten precipitates. From these results, the following general conclusions can be made:

- 1) These precipitates or crystals grow at a constant rate and this rate is constant for a given supercooling (or supersaturation).
- 2) The advancing front of these precipitates appears to be of parabolic shape, and the radius of curvature at the tip of the parabolic interface is smaller than a micron in size.
- 3) The parabolic front preferentially grows along a specific crystallographic direction in a given system. Many theoretical attempts have been made to explain these observations quantitatively. One of the early efforts was by Papapetrov (1) who pointed out the possibility of a constant growth rate if the interface were isothermal and parabolic in shape. Subsequently, Ivantsov (2,3) proved this conclusion quantitatively by obtaining the solution of the heat flow problem, and showed that a constant growth rate would be obtained for an isothermal inter-

face if the interface shape corresponded to a paraboloid of revolution or a parabolic cylinder. From these results, the relationship between the growth rate and supercooling was derived. The result showed that an infinite number of solutions are possible depending upon the radius of curvature,  $\rho$ , at the tip of the parabolic interface. For a given supercooling, growth rates will vary from zero to infinity as the values of  $\rho$  change from infinity to zero. This did not resolve the experimental observation of constant and fixed growth rate at a given supercooling.

The inadequacy of the Ivantsov solution was realized by many investigators (4-6), and was attributed to the assumption of isothermal interface. Zener (4) pointed out that because of the large curvature of the parabolic front, the Gibbs-Thomson effect would be significant so that the interface temperature would be proportional to the curvature of the interface. Subsequently, Temkin (5) and Bolling and Tiller (6) pointed out that non-isothermal interface would also result from the non-uniform interface undercooling required for atomic attachment process at the interface. When non-isothermal interface is considered, the theoretical analysis becomes quite complex since the steady-state interface shape will deviate from a parabola. However, as suggested by Temkin (5), a reasonably valid relationship between growth rates and supercooling will be obtained if one considered a non-isothermal parabolic interface. Temkin (5) obtained an approximate solution of the parabolic interface model, which was subsequently solved rigorously by Trivedi (7-9). Once

the non-isothermal nature of the interface is considered, the growth rate as a function of radius of curvature,  $\rho$ , goes through a maximum for any given undercooling. This maximum growth rate was proposed by Zener (4) to be the one observed experimentally. Theoretical calculations based on the maximum growth rate principle agree well with experimentally observed rate for many systems studied (10).

All the above models have considered a simple case of the growth of an isolated, parabolically shaped, particle in an infinite medium which is initially supercooled to a constant temperature. Most growth conditions, however, are quite complex in that the melt is not supercooled, but has increasing temperature away from the interface. Also, many other parameters such as anisotropy of interfacial energy and of interface kinetic coefficient have been ignored. The dendrites and precipitates usually grow in a parallel array rather than in an isolated manner as assumed. A mathematical solution of a realistic problem becomes quite complex. However, most of these effects can be easily incorporated by appropriately altering the boundary condition of the problem. In order to achieve this, it would be desirable to obtain solutions of heat and mass transfer equations for a general boundary condition, such as that given by a convergent Taylor's series expansion. Two general boundary conditions are considered in this paper, and the solutions of the growth rate as a function of supercooling are obtained for a parabolic cylinder and a paraboloid of revolution shaped interfaces. A specific example is then given to show how these solutions can

be used to theoretically predict the dendrite growth behavior under more realistic growth conditions encountered in crystal growth and in solidification studies of castings.

## 2. Mathematical Description

### 2.1 Basic Differential Equation

Figure 1 shows a schematic diagram of the section of a solid plate or a needle dendrite front growing in a liquid medium. Since these dendrites grow at a constant rate, the temperature distribu-

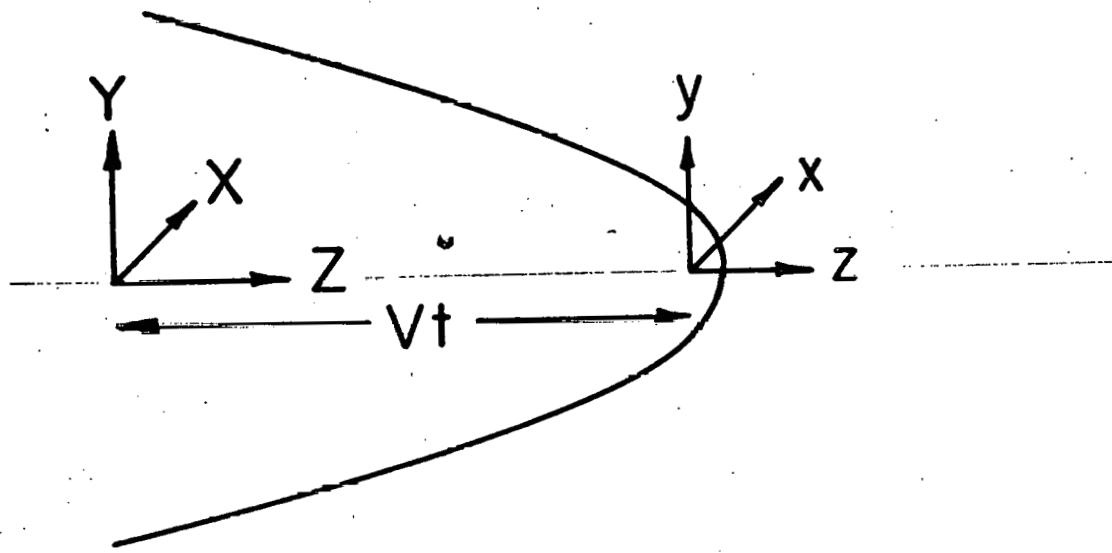


Fig. 1. A schematic diagram of a parabolic interface with an attached moving coordinate system.

tions in liquid and in solid will be governed by the steady-state heat flow equation in a moving coordinate system attached to the interface (11). We shall use dimensionless coordinate system  $(x, y, z) = (X, Y, Z - Vt)/\rho$ , where capital  $X, Y, Z$  refer to a coordinate system fixed in space,  $V$  is the constant growth rate of the dendrite tip, and  $\rho$  the radius of curvature at the growing dendrite tip. The steady-state heat flow is given by (11,12)

$$\nabla^2 T_i + 2p_i \vec{\epsilon} \cdot \nabla T_i = 0 \quad (i=S, L), \quad (1)$$

where subscripts  $S$  and  $L$  refer to solid and liquid regions, respectively.  $p_i$  is the thermal peclet number equal to  $V\rho/2a_i$ ,  $a_i$  being the thermal diffusivity, and  $\vec{\epsilon}$  is a unit vector in the direction of the dendrite tip growth ( $z$  direction in Fig. 1).

Since the shape of dendrite front is parabolic, it will be advantageous to select parabolic coordinate systems. We shall consider both the plate and the needle dendrites. For a needle dendrite, represented by a paraboloid of revolution, we consider a parabolic coordinate system  $(\alpha, \beta)$  in which  $\alpha = 1$  represents the dendritic interface (7). In this system, equation (1) transforms to

$$\frac{\partial^2 T_i}{\partial \alpha^2} + \left(\frac{1}{\alpha} + 2p_i \alpha\right) \frac{\partial T_i}{\partial \alpha} + \frac{\partial^2 T_i}{\partial \beta^2} + \left(\frac{1}{\beta} - 2p_i \beta\right) \frac{\partial T_i}{\partial \beta} = 0 \quad (2)$$

## 2.2 Boundary Conditions

The temperature distribution in liquid far away from the interface ( $\xi$  or  $\alpha \rightarrow \infty$ ) will approach a constant value,  $T_\infty$ . Along the interface the variation of temperature will depend upon many

factors such as interfacial energy, interface mobility, anisotropic nature of interface energy and interface mobility, presence of any solute in the system, etc. However, the temperature profile will always be continuous and finite. Thus, we shall assume a general boundary condition to be given by the expression

$$T_{int} - T_{\infty} = \sum_{m=0}^{\infty} d_m \beta^{2m} \text{ for a needle dendrite} \quad (4)$$

$$\sum_{m=0}^{\infty} d_m \eta^{2m} \text{ for a plate dendrite,} \quad (5)$$

where  $T_{int}$  is the temperature profile along the solid-liquid interface.

Although the above boundary conditions are quite general, the boundary conditions in many cases depend upon physical parameters which vary as a function of  $\cos\theta$ , where  $\theta$  is the angle between the interface normal and the dendrite tip growth direction (7-9). Thus, it is advantageous to obtain a general solution for another boundary condition which is given by a power series in  $\cos\theta$ . We shall therefore consider the following boundary condition

$$T_{int} - T_{\infty} = \sum_{m=0}^{\infty} d_m \cos^{m\theta}$$

or

$$T_{int} - T_{\infty} = \sum_{m=0}^{\infty} \frac{d_m}{(1+\beta^2)^{m/2}} \text{ for a needle dendrite} \quad (6)$$

$$\sum_{m=0}^{\infty} \frac{d_m}{(1+\eta^2)^{m/2}} \text{ for a plate dendrite} \quad (7)$$

The coefficients  $d_m$  in boundary conditions (4-7) are such that the temperature variation along the interface is continuous and finite for all  $\beta$  or  $\eta$ . In the subsequent sections we shall refer to boundary condition (4) or (5) as Type I and boundary condition (6) or (7) as Type II conditions.

### 2.3 Growth Rate Calculations

The solution of the differential equation (1) with boundary conditions (4,5,6 or 7) gives the temperature profiles in solid and in liquid. The relationship between the growth rate and the bath undercooling can then be obtained from the thermal flux balance at the tip of the dendrite. For a needle dendrite the relationship is:

$$\frac{2p_L \Delta H}{C_L} = - \frac{\partial T_L}{\partial \alpha} \Big|_{\alpha=1} + \frac{K_S}{K_L} \frac{\partial T_S}{\partial \alpha} \Big|_{\alpha=1}, \quad (8)$$

where  $C_L$  is the heat capacity of the liquid,  $\Delta H$  the heat of fusion per unit volume, and  $K_S$  and  $K_L$  are thermal conductivities of solid and liquid, respectively. An analogous equation can be written for a plate dendrite, and the result will be the same as equation (8) when the coordinate  $\alpha$  is replaced by the coordinate  $\xi$ . The temperature gradients in liquid and in solid at the dendrite tip can be written in general as

$$-(\partial T_L / \partial \alpha)_{\alpha=1} = \sum_{m=0}^{\infty} d_m F_m \quad \text{and} \quad (\partial T_S / \partial \alpha)_{\alpha=1} = \sum_{m=0}^{\infty} d_m G_m, \quad (9)$$

where the functions  $F_m$  and  $G_m$  are evaluated from the solution of the differential equation. In terms of these functions, equation (8) can be rewritten as follows:

$$2p_L(\Delta H/C_L) = \sum_{m=0}^{\infty} d_m (F_m + \frac{K_S}{K_L} G_m). \quad (10)$$

Our aim in the following part of the paper is to evaluate functions  $F_m$  and  $G_m$  for boundary conditions (4-7).

### 3. Needle Dendrite

#### 3.1 Solution of the Differential Equation

We first consider the solution of equation (2) in the liquid phase by the separation of variables method (7). Assuming a product type of solution

$$T_L \sim f(\alpha) g(\beta),$$

we obtain a pair of differential equations

$$f'' + [(1/\alpha) + 2p_L\alpha]f' - \lambda f = 0, \quad (11)$$

and

$$g' + [(1/\beta) - 2p_L\alpha]g' + \lambda g = 0, \quad (12)$$

where  $\lambda$  is the separation constant, and primes on  $f$  and  $g$  represent differentiation with respect to  $\alpha$  and  $\beta$ , respectively. We first seek the solution of equation (12) which is finite as  $\beta \rightarrow \infty$ . The general solution of equation (12) is given by the confluent hypergeometric function (13).

$$g(\beta) \sim \psi(-\frac{\lambda}{4p_L}, 1, p_L\beta^2)$$

and this solution is finite as  $\beta \rightarrow \infty$  only when  $\lambda/4p_L$  equals zero or a positive integer (14). For these values of  $\lambda/4p_L$ , the hypergeometric function reduces to Laguerre polynomials of zero order, so that the solution of equation (12) is

$$g(\beta) \sim L_n^0(p_L\beta^2) \quad (13)$$

for these values of  $\lambda/4p_L$ , equation (11) has a solution which is finite as  $\alpha \rightarrow \infty$  of the form

$$f(\alpha) \sim \exp(-p_L \alpha^2) \psi(n+1, 1, p_L \alpha^2) \quad (14)$$

The general solution of the differential equation (2) can thus be written as

$$T_L - T_\infty = \sum_{n=0}^{\infty} A_L \frac{\exp(-p_L \alpha^2) \psi(n+1, 1, p_L \alpha^2)}{\exp(-p_L) \psi(n+1, 1, p_L)} L_n^0(p_L \beta^2) \quad (15)$$

The corresponding solution in the solid phase which is finite for  $\alpha = 0$ , is given by

$$T_S - T_\infty = \sum_{n=0}^{\infty} A_S \frac{\exp(-p_S \alpha^2) \phi(n+1, 1, p_S \alpha^2)}{\exp(-p_S) \phi(n+1, 1, p_S)} L_n^0(p_S \beta^2), \quad (16)$$

where functions  $\phi$  and  $\psi$  are confluent hypergeometric functions of first and second kind, respectively. Along the interface  $\alpha = 1$ , the two solutions given by equations (15) and (16) must match. Thus, coefficient  $A_S(p_S) = A_L(p_L)$ . We shall now evaluate the value of this coefficient for the two types of boundary conditions discussed in section 2.2.

### 3.2 Boundary Condition of Type I

Equating equation (14) for  $\alpha = 1$  with the boundary condition (4), we obtain

$$\sum_{n=0}^{\infty} A_L L_n^0(p_L \beta^2) = \sum_{m=0}^{\infty} d_m \beta^{2m}$$

Substituting  $x = p_L \beta^2$  gives

$$\sum_{n=0}^{\infty} A_L L_n^0(x) = \sum_{m=0}^{\infty} d_m p_L^{-m} x^m$$

Expanding  $x^m$  in Laguerre Polynomials (13), and substituting the result in the above expression gives

$$\sum_{n=0}^{\infty} A_L L_n^0(x) = \sum_{m=0}^{\infty} d_m p_L^{-m} \sum_{n=0}^{\infty} \frac{\Gamma(m+1)\Gamma(m+1)(-1)^n}{\Gamma(n+1)\Gamma(m-n+1)} L_n^0(x)$$

Since the temperature profile along the interface is continuous, the value of the coefficient  $A_L$  can be obtained by interchanging the summation orders and comparing the coefficients of  $L_n^0(x)$  on both sides. The result is

$$A_L = \sum_{m=0}^{\infty} \frac{(-1)^n}{n!} \frac{m!m!}{\Gamma(m-n+1)} p_L^{-m} d_m \quad (17)$$

Note that the terms for  $n > m$  are zero.

The values of temperature gradients at the tip of dendrite ( $\alpha=1, \beta=0$ ) in liquid and in solid can now be obtained by differentiating equations (15) and (16) with respect to  $\alpha$ . The result is

$$-\frac{\partial T_L}{\partial \alpha} \Big|_{\substack{\alpha=1 \\ \beta=0}} = 2p_L \sum_{n=0}^{\infty} A_L \frac{\psi(n+1, 2, p_L)}{\psi(n+1, 1, p_L)} \quad (18)$$

and

$$\frac{\partial T_S}{\partial \alpha} \Big|_{\substack{\alpha=1 \\ \beta=0}} = 2p_S \sum_{n=0}^{\infty} A_S n \frac{\phi(n+1, 2, p_S)}{\phi(n+1, 1, p_S)} \quad (19)$$

Substituting the value of the coefficients from equation (16) and comparing the results with equation (9), we obtain the values of the functions  $F_m$  and  $G_m$  as

$$F_m = 2p_L^{-m+1} m! m! \sum_{n=0}^m \frac{(-1)^n}{n!} \frac{1}{\Gamma(m-n+1)} \frac{\psi(n+1, 2, p_L)}{\psi(n+1, 1, p_L)} \quad (20)$$

and

$$G_M = 2p_S^{-m+1} m! m! \sum_{n=0}^m \frac{(-1)^n}{(n-1)!} \frac{1}{\Gamma(m-n+1)} \frac{\phi(n+1, 2, p_S)}{\phi(n+1, 1, p_S)} \quad (21)$$

The specific values of these functions are:

$$F_0 = 2p_L \psi(1, 2, p_L) / \psi(1, 1, p_L) = 2p_L / [p_L e^{p_L} E_1(p_L)]$$

and  $G_0 = 0$ , where  $E_1(p_L)$  is the exponential integral function.

These functions then give the result for the isothermal boundary condition, and the result is identical to that obtained by Ivantsov (2) for this case.

### 3.3 Boundary Condition of Type II

We now evaluate the coefficients  $A_L$  in equation (15) for the boundary condition given by equation (6). At the interface,  $\alpha=1$ , we have

$$\sum_{n=0}^{\infty} A_L L_n^0(p_L \beta^2) = \sum_{m=0}^{\infty} \frac{d_m}{(1+\beta^2)^{m/2}}$$

or

$$\sum_{n=0}^{\infty} A_L L_n^0(x) = \sum_{m=0}^{\infty} \frac{d_m p_L^{m/2}}{(p_L + x)^{m/2}}$$

The coefficient  $A_L$  is then obtained as

$$A_L = \sum_{m=0}^{\infty} d_m p_L^{m/2} \int_0^{\infty} e^{-x} \frac{L_n^0(x)}{(p_L + x)^{m/2}} dx$$

Substituting the integral representation of the Laguerre Polynomials (13), we obtain

$$A_L = \sum_{m=0}^{\infty} d_m p_L^{m/2} \int_0^{\infty} \frac{dx}{(p_L + x)^{m/2}} \int_0^{\infty} \frac{1}{t^n} t^n J_0(2\sqrt{xt}) e^{-t} dt$$

$$= \sum_{m=0}^{\infty} \frac{d_m p_L^{m/2}}{n!} \int_0^{\infty} e^{-t} t^n \int_0^{\infty} \frac{J_0(2\sqrt{xt})}{(p_L + x)^{m/2}} dx dt \quad (22)$$

Substituting  $x/p_L = u^2$  and  $b = 2\sqrt{pt}$ , the integral,  $I$ , inside the large bracket can be written as

$$I = 2p_L^{1-m/2} \int_0^{\infty} \frac{u J_0(bu)}{(1+u^2)^{m/2}} du$$

The solution of the above integral is given by Gradshteyn and Ryzhik (15), and substituting this result in equation (22) gives

$$A_L = \sum_{m=0}^{\infty} \frac{4d_m}{n!} \frac{p_L^{(m+2)/4}}{\Gamma(m/2)} \int_0^{\infty} e^{-\xi^2} \xi^{(4n+m)/2} K_{1-\frac{m}{2}}(2\sqrt{p_L}\xi) d\xi$$

where we have substituted  $\xi = \sqrt{t}$ . The solution of the above integral is also given by Gradshteyn and Ryzhik (22), and the result can be written in terms of confluent hypergeometric function as follows:

$$A_L = \sum_{m=0}^{\infty} \frac{d_m p_L}{\Gamma(m/2)} \Gamma(n + \frac{m}{2}) \psi(n+1, 2 - \frac{m}{2}, p_L) \quad (23)$$

The coefficient  $A_S$  will also be given by equation (23) when  $p_L$  is replaced by  $p_S$ . Substituting the above result in equations (18) and (19), and comparing the result with equation (9) we obtain the values of functions  $F_m$  and  $G_m$  as

$$F_m = \frac{2p_L^2}{\Gamma(m/2)} \sum_{n=0}^{\infty} \Gamma(n + \frac{m}{2}) \psi(n+1, 2 - \frac{m}{2}, p_L) \frac{\psi(n+1, 2, p_L)}{\psi(n+1, 1, p_L)} \quad (24)$$

and

$$G_m = \frac{2p_S^2}{\Gamma(m/2)} \sum_{n=0}^{\infty} n \Gamma(n + \frac{m}{2}) \psi(n+1, 2 - \frac{m}{2}, p_S) \frac{\phi(n+1, 2, p_S)}{\phi(n+1, 1, p_S)} \quad (25)$$

The above result, when substituted in equation (10), correlates the growth of dendritic needles with supercooling when interface temperature varies according to equation (4).

#### 4. Plate Dendrite

The mathematical procedure for the solution of a plate dendrite growth is quite analogous to that for the needle dendrite growth. We shall therefore present the results of the plate dendrite growth in this section.

The differential equation (3) is solved by the separation of variables technique (8,9) and the solution which is finite as  $\eta \rightarrow \infty$  is as follows:

$$T_L - T_\infty = \sum_{n=0}^{\infty} A_L \frac{I_{2n} \operatorname{erfc}(\sqrt{p_L} \xi)}{I_{2n} \operatorname{erfc}(\sqrt{p_L})} H_{2n}(\sqrt{p_L} \eta) \quad (26)$$

and

$$T_S - T_\infty = \sum_{n=0}^{\infty} A_S \frac{\exp(-p_S \xi^2) \phi(n + \frac{1}{2}, \frac{1}{2}, p_S \xi^2)}{\exp(-p_S) \phi(n + \frac{1}{2}, \frac{1}{2}, p_S)} H_{2n}(\sqrt{p_S} \eta) \quad (27)$$

where  $I_{2m} \operatorname{erfc}(x)$  and  $H_{2m}(x)$  are the integral error functions (18) and the Hermite Polynomials (13), respectively.

To evaluate the coefficient  $A_L$  for boundary condition of Type I, given by equation (5), we follow a procedure similar to that for the needle case. The result is

$$A_L = \sum_{m=0}^{\infty} \frac{p_L^{-m} d_m(2m)!}{2^{2m} (2n)! \Gamma(m-n+1)} \quad (28)$$

A similar expression is obtained for the coefficient  $A_S$  when  $p_L$  is replaced by  $p_S$ .

Differentiating equations (26) and (27) with respect to  $\xi$ , and evaluating the gradients at  $\xi=1$ ,  $\eta=0$ , we obtain the values of the function  $F_m$  and  $G_m$  as

$$F_m = \frac{2p_L^{(1/2)-m} (2m)!}{2^{2m}} \sum_{n=0}^m \frac{(-1)^n}{\Gamma(n+\frac{1}{2})} \frac{1}{\Gamma(m-n+1)} \frac{I_{2n-1} \operatorname{erfc}(\sqrt{p_L})}{I_{2n} \operatorname{erfc}(\sqrt{p_L})} \quad (29)$$

and

$$G_m = 4p_S^{1-m} \frac{(2m)!}{2^{2m}} \sum_{n=0}^m \frac{(-1)^n}{\Gamma(n) \Gamma(m-n+1)} \frac{\phi(n+\frac{1}{2}, \frac{3}{2}, p_S)}{\phi(n+\frac{1}{2}, \frac{1}{2}, p_S)} \quad (30)$$

The substitution of equations (29) and (30) in equation (10) gives us the relationship between the growth rate of plate dendrite and bath undercooling.

When boundary condition of Type II, given by equation (7), is considered, the coefficient  $A_L$  is obtained as

$$A_L = \sum_{m=0}^{\infty} \frac{d_m p_L^2}{2^{2m} (2m)! \sqrt{\pi}} \int_{-\infty}^{\infty} \frac{\exp(-x^2) H_{2m}(x)}{(p_L + x^2)^{m/2}} dx,$$

when  $x = \sqrt{p_L} \eta$ . Replacing Hermite Polynomials by its integral representation gives for the coefficient the expression

$$A_L = \frac{(-1)^n}{(2n)!} \frac{4}{\pi} \sum_{m=0}^{\infty} d_m p_L^{m/2} \int_0^{\infty} e^{-t^2} t^{2n} \int_0^{\infty} \frac{\cos 2xt}{(p_L + x^2)^{m/2}} dx dt \quad (31)$$

The solution of the integral with variable  $x$  has been given by Lebedev (13), and substituting the result we obtain for  $m>0$ ,

$$A_L = \frac{(-1)^n}{(2n)!} \frac{4}{\sqrt{\pi}} \sum_{m=1}^{\infty} \frac{d_m p_L^{(m+1)/4}}{\Gamma(m/2)} \int_0^{\infty} e^{-t^2} t^{2n+(m/2)-(1/2)} K_{(m-1)/2}(2\sqrt{p_L} t) dt \quad (32)$$

Substituting  $t^2 = u$ , the integral in equation (32) is of the standard form whose solution is given by Lebedev (18). The coefficient  $A_L$  is then given by the expression

$$A_L = \frac{(-1)^n}{(2n)! \sqrt{\pi}} \sum_{m=1}^{\infty} \frac{d_m p_L^{m/2}}{\Gamma(m/2)} \Gamma(n + \frac{m}{2}) \Gamma(n + \frac{1}{2}) \psi(n + \frac{m}{2}, \frac{m}{2} + \frac{1}{2}, p_L) \quad (33)$$

The equation (32) is valid only when  $m \neq 0$ . The result for  $m = 0$  will be the same as that for the  $m = 0$  case in the Type I boundary condition result.

From the above result, the values of functions  $F_m$  and  $G_m$  can be evaluated as shown before. The results for  $m > 0$  are

$$F_m = \frac{2}{\sqrt{\pi}} \frac{p_L^{(m+1)/2}}{\Gamma(m/2)} \sum_{n=0}^{\infty} \Gamma(n + \frac{m}{2}) \psi(n + \frac{m}{2}, \frac{m}{2} + \frac{1}{2}, p_L) \frac{I_{2n-1} \operatorname{erfc}(\sqrt{p_L})}{I_{2n} \operatorname{erfc}(\sqrt{p_L})} \quad (34)$$

and

$$G_m = \frac{4p_S}{\sqrt{\pi}} \frac{p_S^{m/2}}{\Gamma(m/2)} \sum_{n=0}^{\infty} \frac{\Gamma(n + \frac{m}{2}) \Gamma(n + \frac{1}{2})}{\Gamma(n)} \frac{\phi(m + \frac{1}{2}, \frac{3}{2}, p_S)}{\phi(m + \frac{1}{2}, \frac{1}{2}, p_S)} \psi(n + \frac{m}{2}, \frac{m}{2} + \frac{1}{2}, p_S) \quad (35)$$

## 5. Discussion

All the previous results have been obtained for the growth of dendrites in a one-component system. For the growth of dendrites or precipitates in a binary system, the diffusion of solute must also be considered. The differential equation for the diffusion of atoms is identical to equation (1) when temperature,  $T$ , is replaced by a concentration  $C$ , and the thermal diffusivity,  $a_i$ , is replaced by the diffusion coefficient,  $D_i$ . A flux balance at the interface would then give the relationship between the growth rate and the supersaturation. In general, for dendrite growth in a binary system, both heat and mass transport must be considered simultaneously.

The boundary conditions for the temperature and the composition profiles are not independent, but are related such that the growth rate results based on thermal as well as on solute flux balance at the interface coincide. When such a coupled problem is considered, the boundary conditions for temperature and solute profiles can be shown to follow boundary conditions of Type I and II. The general solutions, derived in this paper, can therefore be used to study such a complex coupled problem under variety of different conditions imposed on the system.

To illustrate the application of the results obtained in this paper, consider a simple case of the growth of needle dendrite in a supercooled melt. Let the interface be isothermal with temperature  $T_M$ , the melting point of the solid. The coefficients in boundary condition (4) are zero except  $d_o$  whose value is  $T_M - T_\infty$ . Equation (1) gives

$$2p_L(\Delta H/C_L) = (T_M - T_\infty)(F_o + \frac{K_S}{K_L} G_o) \quad (36)$$

Equation (21) shows that  $G_o = 0$ , and from equation (20)  $F_o = 2p_L / [p_L e^{\frac{p_L}{T_M}} E_1(p_L)]$ . Substituting these values in equation (36), and rearranging, we obtain

$$(T_M - T_\infty)(C_L / \Delta H) = p_L e^{\frac{p_L}{T_M}} E_1(p_L) \quad (37)$$

The above result is identical to that obtained by Ivantsov (2).

We shall now consider the case where the surface energy and the interface mobility are finite so that the interface temperature

is no longer isothermal but given by the relationship (7)

$$T_{int} - T_{\infty} = (T_M - T_{\infty}) - \frac{\gamma}{\Delta S \rho} \frac{2+\beta^2}{(1+\beta^2)^{3/2}} - \frac{V}{\mu_0} \frac{1}{(1+\beta^2)^{1/2}}$$

where  $\gamma$  is interfacial energy,  $\Delta S$  the entropy of fusion and  $\mu_0$  the interface kinetic coefficient. The above boundary condition can be rewritten as

$$T_{int} - T_{\infty} = (T_M - T_{\infty}) - \left( \frac{V}{\mu_0} + \frac{\gamma}{\Delta S \rho} \right) \frac{1}{(1+\beta^2)^{1/2}} - \frac{\gamma}{\Delta S \rho} \frac{1}{(1+\beta^2)^{3/2}} \quad (38)$$

This boundary condition is of Type II with  $d_0 = T_M - T_{\infty}$ ,  $d_1 = -\left(\frac{V}{\mu_0} + \frac{\gamma}{\Delta S \rho}\right)$  and  $d_2 = -\frac{\gamma}{\Delta S \rho}$ . Equation (10) can therefore be written as

$$2p_L(\Delta H/C_L) = (T_M - T_{\infty})F_0 - \left( \frac{V}{\mu_0} + \frac{\gamma}{\Delta S \rho} \right) \left( F_1 + \frac{K_S}{K_L} G_1 \right) - \frac{\gamma}{\Delta S \rho} \left( F_2 + \frac{K_S}{K_L} G_2 \right) \quad (39)$$

where the functions  $F_m$  and  $G_m$  are given by equations (24) and (25), respectively. Equation (39) can be rearranged as

$$(T_M - T_{\infty})F_0 = 2p_L(\Delta H/C_L) + \frac{V}{\mu_0} \left( F_1 + \frac{K_S}{K_L} G_1 \right) + \frac{\gamma}{\Delta S \rho} \left[ \left( F_1 + F_2 \right) + \frac{K_S}{K_L} (G_1 + G_2) \right] \quad (40)$$

In the above results, we have assumed  $\gamma$  and  $\mu_0$  to be constant along the interface. However, they are usually anisotropic and one may expand them in Tayler's series around  $\beta = 0$ . In this case, the boundary condition of Type I and Type II would give the final result which will show how these anisotropic effects alter the

growth characteristics of dendrites. Similarly, one may profitably use equation (10) to assess how other variables imposed upon the system would alter the growth characteristics of dendrites.

## REFERENCES

1. A. Papapetrov, Z. Krist. 92 (1935) 108.
2. C. P. Ivantsov, Dokl. Akad. Nauk. SSSR 58 (1947) 567.
3. G. P. Ivantsov, Growth of Crystals, vol. 3 (Consultants Bureau, New York, 1960).
4. C. Zener, Trans. AIME. 167 (1946) 550.
5. D. E. Temkin, Dokl. Akad. Nauk. SSSR 132 (1960) 1307.
6. G. F. Bolling and W. A. Tiller, J. Appl. Phys. 32 (1961) 2587.
7. R. Trivedi, Acta Metall. 18 (1970) 287.
8. R. Trivedi, Scripta Metall. 3 (1969) 613.
9. R. Trivedi, Met. Trans. 1 (1970) 921.
10. R. Trivedi and W. A. Tiller, Acta Metall. (in press).
11. D. Rosenthal, Trans. ASME 68 (1946) 849.
12. H. C. Carslaw and J. C. Jaeger, Conduction of Heat in Solids (Oxford University Press, London, 1959).
13. N. N. Lebedev, Special Functions and Their Applications, (Prentice-Hall, Englewood Cliffs, N.J., 1965).
14. P. M. Morse and H. Feshbach, Methods of Theoretical Physics (McGraw-Hill, New York, 1953).
15. I. S. Gradshteyn and I.M. Ryzhik, Table of Integrals, Series, and Products, (Academic Press, New York, 1965).
16. O. S. Berlyand, R. I. Gavrilova and A. P. Prudnikov, Inzh. Fiz. Zhur. 3 (1960) 103.

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