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A mechanism for the equalisation of primary spacing during cellular and dendritic growth

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The Hunt and Lu [*Metall. Mater. Trans. A* **27A** (1996) 611] model for the selection of primary spacing, λ, during cellular and dendritic growth predicts a range of spacings falling between minimum and maximum values of λ_{min} and λ_{max} respectively. Within the model $\lambda_{max}/\lambda_{min}$ will be at least 2 and may, under certain circumstances, be significantly greater. In this paper we use a free boundary model of solidification within an array to demonstrate that interaction between the tips of the cells or dendrites leads to a transverse adjustment mechanism that will tend to equalise the spacing as growth proceeds. This transverse adjustment mechanism is shown to be rapid for the spacings characteristic of cellular growth but much more gradual for the spacings characteristic of dendritic growth. These findings are consistent with observations of the primary spacing of dendrites grown in alloys of the transparent casting analogue, succinonitrile.

1. Introduction

The selection of primary spacing within a cellular or dendritic array during directional solidification has been an enduring problem within the metallurgical literature and is of considerable commercial importance. Early work directed at this problem attempted to find unique relationships between the solidification conditions and the primary spacing, λ, by making certain simplifying assumptions about the array growth problem. By assuming a smooth, steady-state, interface for dendrites in an hexagonal array, in which the temperature and composition of the liquid normal to the principal growth direction was constant, Hunt [1] was able to solve for λ and the interface shape away from the tip region. Kurz & Fisher [2] assumed that a fully developed dendrite, including side branches, could be approximated as an ellipsoid of revolution, whereupon an analysis related to the marginal stability theory for tip radius selection yields expressions for λ in the limits of high or low growth velocity, V. The results thus obtained were qualitatively similar to those of Hunt. The Hunt model was developed further by Trivedi [3] although the agreement between theory and experiment [4] was still incomplete.

An important advance towards the solution of this problem was made by Warren and Langer [5, 6]. They proposed that rather than a unique value of λ there may be an allowable range of primary spacings for any given set of solidification conditions. Following an analysis in which they considered not only the initial growth of instabilities from a planar interface, but also subsequent coarsening, they proposed that the selection of the actual value of λ observed depended not only upon the current conditions of the system but also its solidification history. Moreover, the results obtained were in good agreement with observations [7, 8] based on the transparent analogue system succinonitrile-acetone. However, the Warren & Langer model is unable to account for the ability of a dendritic array to decrease its primary spacing by tip-splitting or higher-order side branching in response to a change in the solidification conditions. This is a phenomenon readily demonstrated in analogue casting systems.

The currently accepted model of primary spacing selection during directional cellular/dendritic growth is due to Hunt & Lu [9]. The Hunt & Lu model, like that of Warren & Langer, proposes that there is an allowable range of values which λ may take. If λ locally falls below its minimum value, λ_{min}, one of the cells or primary dendrites is removed from the array by overgrowth. During this process solute transport from neighbouring cells or dendrites reduces the growth velocity of one the members of the array so that it progressively falls behind the solidification front and is eventually eliminated from the array. Conversely, if λ locally exceeds its maximum value, λ_{max}, a new cell or primary dendrite is nucleated. This occurs by tip splitting in the case of cells or the accelerated growth of a neighbouring tertiary arm for dendrites.

The general validity of the Hunt & Lu model has been demonstrated by a number of studies using analogue casting systems. In dendritic systems, Losert *et al.* [10] found that in directionally solidified succinonitrile-C152, reduction of the solidification velocity leads to a period doubling instability. Within their array of uniformly spaced dendrites, alternate dendrites

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progressively lost their side branches before being eliminated from the array by overgrowth. The growth of new primary dendrites from tertiary arms has been demonstrated [11] in a succinonitrile-ethanol alloy subjected to an increasing solidification velocity. In cellular systems changes in primary spacing by both overgrowth and tip splitting have been observed in succinonitrile-acetone by Han & Trivedi [12] and in succinonitrile-C152 by Losert *et al.* [13]. Measured primary spacings derived from these experiments are generally in good accord with the model of Hunt & Lu.

However, the variation in the primary spacing predicted by the Hunt & Lu model is quite large. Clearly the ratio $\lambda_{max}/\lambda_{min}$ must be at least 2. If this were not the case the elimination of a cell or dendrite from the array could drive the local spacing above λ_{max} leading to the eliminated feature being immediately replaced and *vice versa*. The results of Losert *et al.* [10] would tend to suggest that in many situations the actual value of $\lambda_{max}/\lambda_{min}$ is very close to 2. This though is inconsistent with the rather uniform primary spacing frequently observed in analogue casting experiments. Recently Ding *et al.* [14] have identified two distinct mechanisms which contribute to the primary spacing in succinonitrile-ethanol and succinonitrile-acetone alloys. One is competitive growth between neighbouring dendrites of the type described by Hunt & Lu. The other is a relatively slow, transverse adjustment driving the spacing towards greater uniformity. The former mechanism determines the mean primary spacing, $\bar{\lambda}$, while the secondary transverse adjustment mechanism leaves $\bar{\lambda}$ unaltered but reduce the standard deviation in λ. Moreover, this transverse adjustment appears to be velocity dependent. Measurements by Hunt *et al.* [15] on the succinonitrile-acetone systems have shown that as the growth velocity increases the array becomes more irregular, that is there appears to be less transverse adjustment. Highly irregular array spacing as a result of rapid solidification has also been observed by Pan *et al.* [16] is laser remelted Cu-Mn alloy.

As a cell or dendrite grows, heat and solute are reject from the tip into the undercooled melt ahead. In the case of an isolated, parabolic, dendrite the isotherms (or isocontrate lines) will form a family of concentric paraboli around the freezing front. For a regular array of cells or dendrites with uniform spacing, λ, the isotherms will no longer be concentric paraboli but will be symmetric about the tip. However, if we now consider an array with non-uniform spacing the isotherms will no longer be symmetric about the tips. Consider a dendrite with its two nearest neighbours at spacings of λ_1 and λ_2, such that $\lambda_1 < \lambda_2$. Due to the closer proximity of the dendrite with spacing λ_1 the isotherms will be more tightly packed towards the dendrite at λ_1 than they are for the dendrite at λ_2. Consequently, as the dendrite grows, with the tip following the isotherms, the dendrite will bend towards its neighbour at λ_2, tending to equalise the array spacing. We would stress that while the overgrowth mechanism of Hunt & Lu implies that there is a transverse movement of the cell or dendrite tips, this motion is assumed rather than calculated and the rate at which it occurs is not determined. It is this transverse adjustment mechanism with which we are concerned in this paper.

2. A free boundary model of growth within an array

In order to quantify this effect a free boundary model of growth within an array [17] has been used. This is a thermal solidification model developed originally to study ripening within a dendritic array. Holding the boundary ahead of the array at a fixed temperature such that solidification is always into a slightly undercooled melt ensures directional dendritic growth. The computational procedure is also valid for pure solutal growth and the results may also have interesting consequences for directional solidification in alloys.

Growth of the dendrite into its parent melt is controlled by the diffusion equation

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T \tag{1}$$

where α is the thermal diffusivity. At the solid/liquid interface growth will occur at velocity v along the local outward pointing normal, $\hat{n}$, subject to the balance of heat fluxes

$$H\rho v = \kappa_s G_s - \kappa_1 G_1 \tag{2}$$

where ρ is the density, taken here as being the same in the solid and liquid states, H is the latent heat of fusion, κ_s and κ_1 are the thermal conductivities in the solid and liquid respectively, and G_s and G_1 are the thermal gradients at the interface along $\hat{n}$ in the solid and the liquid.

The interface temperature is fixed by its geometry. For a solid growing with an anisotropic, four-fold symmetric interfacial energy γ, the local interface liquidus temperature, T_i, is given by

$$T_i = T_m - \frac{K\gamma_o T_m}{H\rho}\{1 - a\cos(4\phi)\} \tag{3}$$

where γ_o is the nominal interfacial energy between the solid and liquid phases, a the surface energy anisotropy, K the surface curvature, ϕ the angle between $\hat{n}$ and the principal growth direction and T_m is the equilibrium liquidus temperature.

Solutions to the diffusion problem are sought on a regular, 2-dimensional M × N grid using a locally one dimensional (LOD) finite difference scheme [18] to yield the temperature at the advanced time step.

In order to simulate solidification and melting the model independently tracks the solid fraction, p, at each grid point. p takes values $0 \le p \le 1$, where $p = 1$ denotes the material being fully solid and $p = 0$ fully liquid. If at any node $0 < p < 1$ the volume cell which has that node as its centroid will contain some part of the freezing front and the temperature at that node is fixed at the local liquidus temperature. At the end of each time step p is updated at each node for which $0 < p < 1$ by considering the heat flux into or out of the volume element during the time step.

For volume elements containing the solidification front, p will take values $0 < p < 1$. However, unlike the

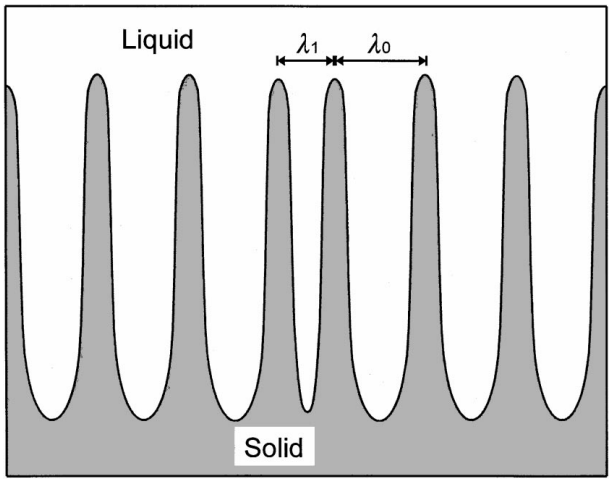

Figure 1 Geometry of the cellular/dendritic array used in the free boundary model.

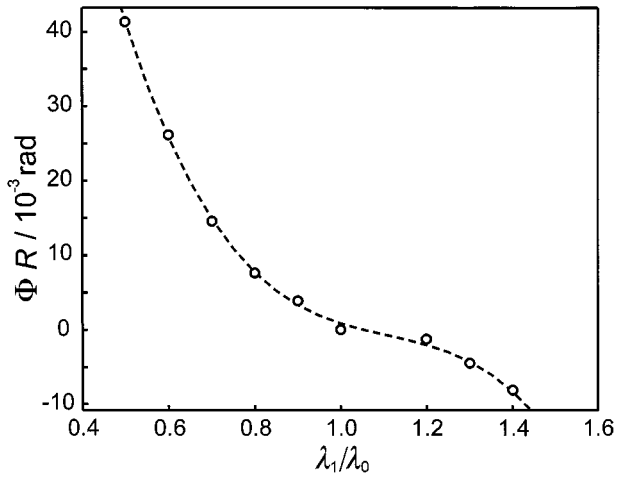

Figure 2 The tip rotation parameter Φ, normalised against the tip radius R, as a function of λ_1/λ_0, for $\lambda_0 = 7.4R$.

phase field approach pioneered by Kobayashi [19], in which the solidification front is assumed diffuse, here we assume the solidification front is sharp and must have a definite position. Consequently we interpret a value of $0 < p < 1$ as meaning the volume cell which has the node (m, n) as its centroid contains some part of the front such that

$$p_{m,n} = \frac{\int_{x_m - \delta x/2}^{x_m + \delta x/2} y \, dx}{\delta x \delta z} \tag{4}$$

where y is the locus of the solidification front and δx & δz are the grid spacing in the x- & z-directions. Full details of the computational procedure are given by Mullis [17, 20].

To determine the effect of non-uniform primary spacing on growth we have considered an array of six dendrites growing in the $+z$ direction (Fig. 1). The primary spacing between the two central dendrites is λ_1, the primary spacing between all other dendrites being λ_0. Within the model the high z boundary is held at a constant undercooling ΔT with the other three boundaries being adiabatic. After an initial period of thermal equilibration, in which the freezing front is fixed in space, the solid-liquid boundary is allowed to move freely under the influence of the thermal field towards the undercooled boundary. During the growth the x & z co-ordinates of the two central dendrite tips are tracked as a function of time to establish if any self-adjustment of the initial spacing occurs.

3. Results

Due to the computationally intensive nature of the free boundary calculation, the model has only been run for times such that the deflection of the tip, Δx, is small relative to the initial spacings λ_0 and λ_1. Under these conditions we find that for all the simulations run

$$\Delta x \propto \Delta z^2 \tag{5}$$

A similar result was found by Mullis [20] for the bending of dendrites growing in a shear flow. For growth at constant velocity, V, this was shown to be consistent with rotation of the principal growth direction at a rate Φ per unit length.

As this deflection is due to the interaction of the thermal fields at the cell or dendrite tips, we might expect that it would be controlled by the extent to which the fields overlap. Consequently, we have parameterised Φ in terms of the variables λ_1/λ_0, λ_0 and P_t, where P_t is the thermal Peclet number in the liquid

$$P_t = \frac{VR}{2\alpha} \tag{6}$$

and R is the radius of curvature at the tip. That the interaction depends upon the magnitude of λ_0 (or λ_1) is clear from the limiting case of very large spacing ($\lambda \to \infty$) when each dendrite will grow essentially independently of its neighbours irrespective of the relative spacing. A dependence upon P_t might be expected because the spatial extent of the thermal field decreasing more rapidly with P_t than the tip radius and consequently as P_t increases the thermal field becomes more localised.

Fig. 2 shows the bending parameter, Φ as a function of the ratio λ_1/λ_0. Here Φ is normalised against the tip radius, ΦR giving the rotation per distance R. $\Phi > 0$ is defined as indicating that the two cells or dendrites are bending away from each other (λ_1 increasing) and $\Phi < 0$ as indicating that the two cells are bending towards each other. In these simulations we have fixed $\lambda_0 = 7.4R$ and $P_t = 1.14 \times 10^{-4}$. It can be seen from the figure that the curve is relatively flat in the vicinity of $\lambda_1/\lambda_0 = 1$ but that Φ increase steeply as λ_1/λ_0 departs significantly from 1, particularly for the case in which the two features become close together. By definition $\Phi = 0$ for $\lambda_1/\lambda_0 = 1$.

The results of varying λ_0 whilst fixing the ratio $\lambda_1/\lambda_0 = 0.7$ are shown in Fig. 3, again with P_t fixed at 1.14×10^{-4}. From the figure it is apparent that the bending parameter, Φ, decreases rapidly as the nominal spacing, λ_0, increases. The results are consistent with Φ decreasing exponentially with λ_0, although this is difficult to verify at large λ_0 due to the very small deflection, Δx, involved.

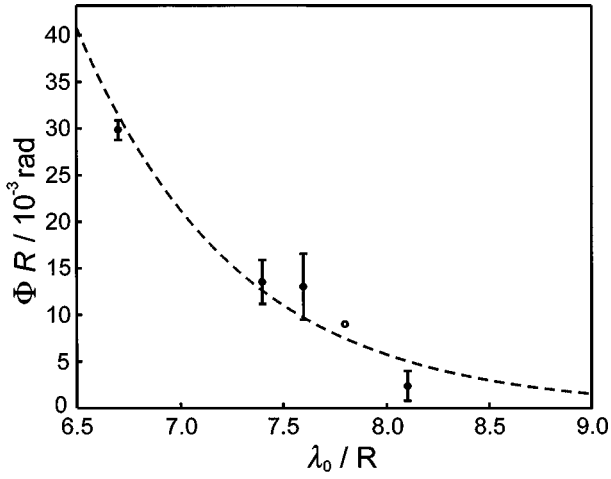

Figure 3 The tip rotation parameter Φ, normalised against the tip radius R, as a function of λ_0, for $\lambda_1/\lambda_0 = 0.7$.

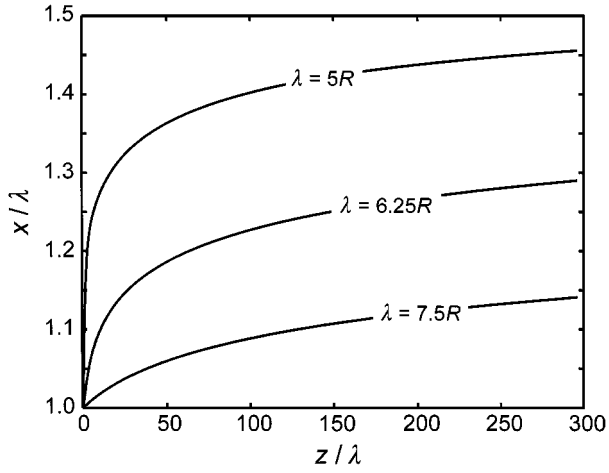

Figure 5 Example calculation showing the locus of a dendrite/cell tip with nearest neighbours at $x = \lambda$ and $x = 3\lambda$.

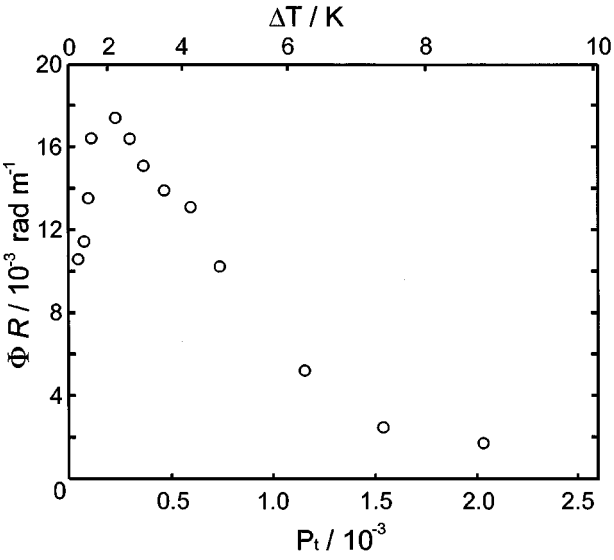

Figure 4 The tip rotation parameter Φ, normalised against the tip radius R, as a function of the thermal Peclet number in the liquid, P_t. (Spacing fixed with $\lambda_0 = 7.4R$ & $\lambda_1/\lambda_0 = 0.7$).

Finally, we have investigated the dependence of Φ upon P_t, the results of which are shown in Fig. 4. The array spacing is fixed with $\lambda_0 = 7.4R$ and $\lambda_1/\lambda_0 = 0.7$. Over most of the range of P_t studied the behaviour is as expected, with the bending parameter Φ decreasing with increasing P_t. However, for $P_t \leq 2.26 \times 10^{-4}$ the opposite trend is observed with Φ increasing with increasing P_t, giving rise to a local maximum in Φ in the vicinity of $P_t = 2.26 \times 10^{-4}$. Thus for a given Peclet number there is an optimum spacing which will maximise the rate at which the dendrite tips bend away from each other. This appears to be related to the point at which the thermal fields from neighbouring dendrites interact. In the case of very low Peclet numbers the thermal fields overlap to such an extent that the isotherms ahead of the solidification front are almost planar and the tendency towards equalisation of the spacing is reduced.

In order to illustrate the effect of the calculated bending rates on the evolution of a cellular or dendritic array we consider the following simple example. Consider three cells/dendrites which at $t = 0$ have tip positions at $x = 0$, $x = \lambda$ and $x = 3\lambda$, so that within this 2-dimensional array the primary spacings for the feature at $x = \lambda$ are $\lambda_{min} = \lambda$ and $\lambda_{max} = 2\lambda$. This gives the ratio $\lambda_{max}/\lambda_{min} = 2$, which according to the results of Losert *et al.* [10] may be close to the maximum value for this parameter. Due to interactions between neighbouring features there will be a tendency for the spacing to adjust to reduce the value of $\lambda_{max}/\lambda_{min}$. Using the results presented in Figs 2 & 3 we may calculate the rate at which the growth direction changes as a result of the interactions between the dendrites and hence calculate the locus that the growing dendrite will follow. The result of this calculation is shown in Fig. 5, for three values of λ, $5R$, $6.25R$ and $7.5R$. The calculation is performed to first order only. We consider only nearest neighbour interactions, which may not be the case in the free boundary model, particularly in the cellular case. Moreover, we do not consider translation of the two features at $x = 0$ and $x = 3\lambda$, which would only be the case if these features were externally constrained. Nonetheless we consider that this is a useful calculation to illustrate the general nature of the process of spacing equalisation.

It is apparent from Fig. 5 that for $\lambda \leq 5R$, which is the case for cells, the adjustment of the primary spacing is very rapid, particularly during the early stages of the process when $\lambda_{max}/\lambda_{min}$ is high. However, it is apparent that as λ_{min} is increased the mechanism rapidly becomes less efficient, with virtually no reduction in $\lambda_{max}/\lambda_{min}$ for $\lambda_{min} \geq 7.5R$. Consequently, for dendritic, as opposed to cellular, systems the adjustment mechanism is very slow. This is a consequence the rapid decrease in Φ with λ_0, illustrated in Fig. 3.

4. Summary & conclusions

Using a free boundary model of solidification we have demonstrated that there is an interaction between the tips of the cells or dendrites growing in an array. This interaction is such that as growth proceeds, it will tend to equalise the primary spacing. This effect has been quantified using a free boundary model of array

solidification. This model suggests that transverse adjustment would be very rapid for the spacings characteristic of cellular growth but much more gradual for the larger spacings (relative to tip radius) characteristic of dendritic growth. These findings are consistent with the observations of Ding *et al.* [14] who report observing such a gradual transverse adjustment of the primary spacing of dendrites grown in transparent succinonitrile-ethanol and succinonitrile-acetone alloys.

The tendency towards equalisation of the primary spacing is also dependent upon the Peclet number in the liquid. It is likely that during rapid solidification (undercooling at the tip >10 K) the thermal field is effectively localised to the dendrite and little transverse adjustment is effected. This observation appears to be consistent with observations in both succinonitrile-acetone alloys and laser melted Cu-Mn alloy.

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