Microstructure development in the directionally solidified
Al–4.0 wt% Cu alloy system

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Abstract

The effect of convection on microstructure formation is examined experimentally and theoretically for the vertically upwards-directional solidification of Al–4.0 wt% Cu alloys. In this alloy system, the rejected solute is heavier than the solvent so that fluid flow occurs due to the presence of radial gradients in temperature and composition. A numerical model is presented which shows that convection effects cause the composition to vary along the interface such that the composition increases from the center to the periphery of the sample. This composition variation causes the macroscopic interface to become convex, and give rise to a systematic variation in microstructure along the interface. Critical experiments have been carried out to examine planar to cellular (and cellular to dendritic) transition in a given sample due to the increase in concentration along the interface, and the experimental results are analyzed through the measurements of interface composition and thermal gradient. In addition, the variation in local primary spacing with interface composition is also characterized and compared with the results of the numerical model. It is shown that microstructure transitions and microstructural scales can be correlated quantitatively with the theoretical results based on interface composition and on temperature and solute gradients at the interface. © 2001 Published by Elsevier Science Ltd.

Keywords: Al–40 wt% Cu alloy system; Microstructure; Numerical model

1. Introduction

Directional solidification (DS) in both metallic and transparent organic systems has been conducted extensively to investigate the formation and evolution of solidification microstructures because the process variables (alloy composition, $C_0$, growth velocity, $V$ and temperature gradient, $G$) can be independently and precisely controlled. Depending on the specific solidification conditions ($C_0$, $V$ and $G$), liquid/solid (L/S) interface could be planar, cellular or dendritic. Experimental studies have been concentrated on establishing relationships between the length scales of solidification microstructures and the externally controlled parameters. These length scales, to a large extent, control the final properties (mechanical, electromagnetic, optical, etc.) of the solidified products made by processes such as casting, welding, functional crystal growth, and laser surface treatment (modification).

The majority of the prevailing theoretical models are based on the diffusive transport of heat and mass transfer during the solidification processes, e.g. the Mullins–Sekerka planar interface stability theory [1], the steady-state dendrite growth model [2], and primary/secondary arm spacing selection [3,4]. With well-controlled experimental conditions in thin samples and precisely determined thermophysical properties of the experimental medium, these models have been reasonably well substantiated. However, in bulk metallic samples, convection is always present in the solidification/crystal growth processes conducted terrestrially. The presence of convection significantly alters the microstructure and microstructure scales so that it is not possible to interpret the microstructure in terms of the diffusive models only.

The nature of convection depends upon the relative orientations of the gravitational vector and the growth direction. For the horizontal DS of transparent systems, it was shown clearly by using the tracer particles that melt flow exists in the liquid driven by horizontal temperature gradient, and that the flow rate is around $20 \mu m s^{-1}$ within a thin sample of $\sim 350 \mu m$ [5]. For the vertically upward solidification process, the mode of convection depends upon whether the solute is lighter or heavier than the solvent. If the solute is lighter than the solvent (in systems such as

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SCN–Ethanol, Pb–Sn, NH₄Cl–H₂O and superalloys for turbine blades), the solute-enriched region in the mushy zone and/or in front of the growth interface can destabilize the bulk liquid and give rise to double diffusive convection [6]. On the other hand, if the solute is denser than the solvent (in systems such as Al–Cu, Sn–Cd, Pb–Au and Sn–Pb), it is generally believed that no convection occurs since both the temperature and composition gradients have a stabilizing effect on the density stratification. In Bridgman growth, a radial temperature difference is always present due to the mismatch of boundary conditions at the insulator-cold zone and insulator-hot zone, and due to the difference in the thermal conductivities of the solidifying alloy and the containing ampoule [7–11]. The melt near the ampoule is hotter than that in the center, so it tends to rise. With horizontal temperature or density gradients, there is no threshold for convection, so fluid motion will always be present, however small the radial gradient is. The intensity of flow, however, can be large or small, and depends on the Rayleigh number, given by:

$$Ra = \frac{g \beta \Delta T d^3}{v \alpha}$$

where g is the gravitational vector, β the thermal expansion coefficient, ΔT the radial temperature difference (generally several degrees), d the diameter of the ampoule, v the kinematic viscosity of the melt, and α the thermal diffusivity of the melt. The related properties for Al–4.0 wt% Cu alloy are listed in Table 1. Numerical calculations by Kim et al. [7,8] and Mazumder et al. [9–12] have established clearly that significant convection due to radial temperature gradient can occur in vertical Bridgman systems even with axially stabilizing temperature and solute gradients (the rejected solute is more dense than the solvent).

The effect of convection on microstructure depends on the mode and intensity of convection. In systems with heavier solute, the intensity of flow is usually weak, but the small variation in composition along the interface significantly influences the macroscopic shape of the interface, and gives rise to a systematic change in microstructure characteristics from the center to the periphery of the sample. In fact it will be shown in this paper that this systematic variation in composition along the interface can be used advantageously to study the effect of composition on microstructures formed under the same conditions of G and V in a given sample.

The main aim of this paper is to address the effect of convection, driven by the radial temperature gradient, on the interface morphology of directionally solidified Al–4.0 wt% Cu alloy. A new experimental method designed for systematic study on the effect of convection is first introduced, in which samples of different dimensions are solidified under exactly the same growth conditions (C₀, V and G). This technique also has an added advantage that in very fine samples, convection in the melt can be minimized so that diffusive growth conditions can be achieved even in ground-based experiments. Next, experimental observations are presented on the effect of different extents of convection on microstructural scales and microstructural transitions. Finally, a numerical model of convection will be presented which will characterize the composition variation along the interface in the Al–Cu system.

The effect of fluid flow in the melt is to alter the concentration profile in the liquid, and thus to change the composition of solute at the interface. The present experimental results show that the actual microstructure that forms under convective conditions in the Al–Cu system can be described by local conditions of interface composition, concentration gradient and thermal gradient in the liquid at the interface.

### 2. Experimental procedure

A conventional Bridgman-type DS apparatus, with a thermal assembly and a translation system, was used in this study. The thermal assembly consists of a tube furnace and a cooling chamber containing liquid Ga–In–Sn eutectic alloy as the cooling medium. The sample was placed in a 600 mm long alumina tube (as ampoule) of ID = 5.0 or 5.5 mm. The ampoule was first evacuated to remove oxygen and other gaseous elements, and then filled with inert Ar. The furnace temperature was raised to the required value for melting the alloy and also establishing the desired temperature gradient. The temperature gradient used in the present study ranged from 6.0 to 7.0 K mm⁻¹, though even a higher gradient could be achieved. Next the thermal assembly was translated vertically upwards at constant speed that spanned from 0.2 µm s⁻¹ to several millimeters per second. The combination of G and V covers the range from planar to cellular to dendritic growth morphology.

Since natural convection in the melt depends strongly on the Rayleigh number, which is a function of tube diameter, reduction in the size of the latter greatly weakens the melt flow. Rigorous numerical calculation of thermo-solutal convection in the Al–Cu system clearly indicates that

| Properties                  | Magnitude   |
|-----------------------------|-------------|
| Melting temperature (T_m, K)| 922         |
| Thermal diffusivity in liquid (α, cm² s⁻¹) | 0.37        |
| Kinematic viscosity (ν, cm² s⁻¹) | 2.6 × 10⁻³  |
| Solute diffusivity in liquid (D_s, cm² s⁻¹) | 5.0 × 10⁻⁵  |
| Thermal expansion coefficient of liquid (β_t, K⁻¹) | 1.0 × 10⁻⁴  |
| Solutal expansion coefficient of liquid (β_s, wt%⁻¹) | 9.2 × 10⁻³  |
| Prandtl number (Pr = ν/α) | 0.007       |
| Schmidt number (Sc = ν/D_s) | 52.0        |
| Liquidus slope (m_L, K wt%⁻¹) | −3.73       |
| Solute partition coefficient (k) | 0.17        |
| Gibbs–Thompson coefficient (Γ, K cm) | 1.2 × 10⁻⁵ |
attributed to the sample size effect, or the natural convection in the melt. The post-experiment analysis of the solidified samples involved microstructure characterization using optical microscopy and composition measurements using SEM-EDs.

3. Experimental results

3.1. Macroscopic L/S interface shape

It is generally assumed that heat flow in a DS process is unidirectional, i.e. perpendicular to L/S interface, and that macroscopic L/S interface shape is flat. However in a common design of a DS unit with radial heating and cooling arrangement, a radial temperature gradient always exists that can lead to a curved L/S interface.

Fig. 2 shows the quenched L/S interfaces in Al–4.0 wt% Cu samples of different diameters. The shape of the interface in sample size of φ5.0 mm is shown in Fig. 2(a) which shows clearly that the interface is highly curved and convex. Fig. 2(b) and (c) show the interface shapes in reduced sample sizes, i.e. 1.0 and 0.6 mm, respectively, and the interface convexity is observed to decrease correspondingly. All other samples except the largest one (φ5.0 mm) were prepared with the thin tube bundle immersion technique described in Section 2.

A parameter, δ, is defined as the distance from the highest point of the interface to the line connecting the two end points of the interface at the ampoule wall, and is taken as a measure of the deviation of the interface shape from a flat interface. This is similar to the definition of the parameter of deflection by Kim and Brown [7]. All of the measurements were made on the previous microstructures (Fig. 2), the results being plotted in Fig. 3, and showing that the interface becomes more convex as the sample diameter increases.

The interface convexity also depends on the morphology of the interface. For a cellular or dendritic interface, the macroscopic curvature of the interface is not so large, as shown in Fig. 4. Except in the periphery region, the macroscopic L/S interface is almost flat. The main difference between a columnar crystal front (dendrite or cell) and a planar front growth lies in the fact that the former can also diffuse the rejected solute into intercellular or interdendritic regions. Concisely speaking, columnar crystals can trap solute between them, known as micro-segregation, and this locally redistributed solute (within the range of primary spacing) tends to lower the composition variation along the macroscopic interface and thus flatten the solidification interface.

3.2. Mixed structures on a growth front

Since convection effects give a non-uniform microstructure over the entire cross-section of a sample, experiments were designed near the planar to cellular and cellular to dendritic transitions that would produce mixed
microstructures in a single sample. Fig. 5(a) shows a sample that is planar in the central region, but cellular at the periphery. Certainly, somewhere close to the ampoule wall, there is a planar-to-cellular transition. Similarly, Fig. 6(a) gives the cellular/dendritic co-existence structure in one sample. Cells occupy most of the cross-section, but near to the periphery are dendrites. Since $G$ and $V$ are the same in a given experiment, these studies provide valuable information on the interface morphologies developed due to the variation in interface composition.

To understand the formation of a mixed microstructure, the compositions in the liquid at a growing interface were determined across the sample, the results being shown in Figs. 5(b) and 6(b). A general tendency in both samples was
that the composition continually increased from the center to the periphery, i.e. radial segregation occurred, which is consistent with the mode of convection due to radial temperature gradient. From these composition profiles, it is not difficult to understand why the periphery is unstable compared with the central regions of the sample. Since the local composition is higher at the periphery than that at the center, a planar interface becomes unstable when the interface composition increases beyond its threshold value.

The mixed microstructures are sensitive to the growth conditions. From the modified supercooling approximation of the Mullins–Sekerka stability analysis [1], one can obtain the critical $G/V$ ratio for planar front stability for Al–4.0 wt% Cu sample as:

$$\frac{-G}{V_{\text{critical}}} = \frac{mC_p(1 - k)}{kD} = 14.57,$$

where $V$ is in $\mu$m s$^{-1}$ and $G$ is the thermal conductivity.

Fig. 4. Quenched cellular morphology ($\times 30$, $G = 6.5$ K mm$^{-1}$, $V = 7.2$ $\mu$m s$^{-1}$). The L/S interface is rather flat except at the periphery.

Fig. 5. Showing: (a) mixed planar/cellular interface morphology ($G = 6.5$ K mm$^{-1}$, $V = 0.45$ $\mu$m s$^{-1}$); (b) measured interface composition as a function of radial position. Superimposed in the figure is the further calculation of interface undercooling parameter $mG_i - G_L$. 
averaged temperature gradient across the interface (K mm\(^{-1}\)). \(G = (k_s G_s + k_l G_l)/(k_l + k_s)\), in which \(k_s\) and \(K_l\) are temperature gradients on the solid and liquid side of the interface. Interface thermal flux balance requires: \(k_s G_s - k_l G_l = V \Delta H\), where \(\Delta H\) is the volumetric heat of fusion. Because \(V \Delta H\) is much smaller than \(k_l G_l\) (or \(k_s G_s\)) at low growth velocities, then \(k_s G_s \approx k_l G_l\). With \(k_s \approx 2k_l\) (for aluminum alloys), one can obtain \(G = (2/3)G_l\), so the critical ratio for the planar front to be stable is:

\[
(G_l/V)_{\text{critical}} = 21.86.
\]

From Fig. 5(a), \(G_l = 6.5 \text{ K mm}^{-1}\) and \(V = 0.45 \mu\text{m s}^{-1}\), so \(G_l/V = 14.44\), which is smaller than the critical value for a stable planar front so that the overall cross-section should be cellular. However, in this large sample (\(\phi 5.5 \text{ mm}\)), convection effects are present which enhanced the planar interface stability [13]. Thus, the center was planar, whereas the periphery was cellular due to increased solute concentration. Slight deviations from this \(G_l/V\) value would lead to either a complete planar front or a complete cellular structure.

3.3. Primary arm spacing

In the presence of convection, primary arm spacing was
also found to be non-uniform across the sample. Fig. 7(a) shows a cross-section of the sample solidified under the same conditions as in Fig. 6(a) where a mixed cellular and dendritic structure formed.

The primary arm spacing at the periphery was larger than that at the center. Fig. 7(b) gives the variation of primary spacing across the interface, indicating clearly a continual increase away from the center. The variation in primary spacing with distance can be correlated to the variation in tip composition with distance, as shown in Fig. 7(b).

Primary spacing were measured by first dividing the cross-section into five co-axial shells (the inner-most one was a circle) at diameters equal to 1.0, 2.0, 3.0, 4.0, and 5.5 mm. The number of primaries in a shell was then counted and designated as $N$. Taking the area of each shell, $A$, and the number $N$, the average area each primary occupied was $A/N$. By assuming a hexagonal arrangement of the primaries and taking the primary arm spacing as $\lambda_1$, it was found that the area that a dendrite should take could be expressed as: $(3/2)\lambda_1^2 \tan 30 = 0.866\lambda_1^2$. At last letting $0.866\lambda_1^2 = A/N$, $\lambda_1$ could be calculated. This is also shown in Table 2 in detail.

4. Discussion

In this part, a numerical model on convection effects in the Al–Cu system will be presented first, then a
through discussion of the above experimental results will be given.

### 4.1. Numerical model

A full-scale numerical model was developed that included: thermo-solutal convection and solute transport in the melt; and heat transfer in the ampoule, crystal and melt that are surrounded by a moving thermal assembly which consists of hot and cold zones separated by an adiabatic region. Presented here are only results that are germane to the Al–Cu system; the details of the model are given in [9,10,14]. One main feature of the application of this model is that the directly measured temperature profiles in both the sample and the furnace used in the present experiments were used to obtain the required value of the Biot number or heat transfer coefficient that is needed to realistically model the interaction between the furnace and the ampoule. Thus, this model specifically applies to the present furnace configuration only, although similar measurements can be made for other Bridgman systems. The main results of the model for DS of Al–4.0 wt% Cu alloy will now be described.

1. There is a radial temperature gradient across the sample, with the temperature at the periphery being 1.0–5.0°C higher than that at the center, depending on the furnace temperature settings. This is a pertinent feature related with the design of the thermal assembly in a Bridgman furnace, as has been analyzed by Trivedi et al. [11] and Favier [15].

This temperature difference contributes in part to the macroscopic convex shape of the L/S interface. A static experiment was conducted to check this effect. It was an experiment in which a sample was kept stationary for a long time before being quenched. The thermal field inside such a sample could be calculated easily. With this thermal distribution and the Al–Cu phase diagram, the L/S interface shape can be determined. The calculated interface shape is shown in Fig. 8. The quenched interface is also superimposed in the figure, indicating the good agreement between the calculated and experimentally observed static L/S interface except at the extreme periphery region. Therefore the interface convexity of the interface in a static process could be well understood through this thermal effect. This is very beneficial in separating other factors responsible for the non-planarity of the L/S interface.

![Fig. 8. Calculated L/S interface shape and its superimposition with the quenched interface of a sample. The modeling condition and the growth conditions are the same: hot zone temperature 800°C; cold zone temperature 22°C; translation velocity zero; sample size 5.0 mm; alumina tube ampoule with a wall thickness of 1.5 mm).](image-url)
is also verified by the experiments, as seen in Fig. 2. The interface is rather flat for a φ0.6 mm sample, but highly convex in the φ5.0 mm sample.

In a bulk sample (e.g. φ5.0 mm), convection dominated the mass transfer in the liquid and the flow just ahead of the interface was laminar in nature in Al–4 wt% Cu alloy and parallel to the interface. The parallel shear flow enhances L/S interface stability, as has been proposed by Delves [13] and experimentally observed by Huang et al. [16].

4.2. Interface morphology

4.2.1. Interface stability

With the melt flow in a bulk sample, one should be cautious in defining the interface stability, since the periphery region contains more solute than the central region and is therefore more prone to instability. The microstructure in Fig. 5(a) shows this scenario clearly. With the measured interface composition, one can check the interface stability locally, at every point of the interface. This has been done for the microstructures shown in Figs. 5(a) and 6(a).

The method for stability analysis is to find the interface constitutional undercooling \( mG_c - G \), where \( G_c \) is the concentration gradient at the interface) through the measured interface composition, \( G_i \), and using the flux balance at the interface:

\[
VC_i(1 - k) = -DG_c
\]

Calculations for Fig. 6(a) are given in Fig. 6(b). It can be seen that \( mG_c - G \) along the interface varies from negative (no constitutional undercooling, so the planar front is stable) to positive (with constitutional supercooling, thus the interface destabilized into cells). This agrees with the present experimental observations, and shows that local conditions can be used to characterize microstructural selection in the Al–Cu system.

A similar reasoning can also be applied to the microstructure transition in Fig. 6(b) for the cell/dendrite transition. From cellular growth, the supercooling \( mG_c - G \) at the tip should be small due to small capillarity undercooling or larger tip radii values. However, for cells near to the cell–dendrite transition, the tip curvature is significant so that a finite value of constitution supercooling will be present to offset the curvature contribution. For dendritic growth, the supercooling \( mG_c - G \) at the tip should be balanced by the stabilizing effect from capillarity at the tip, i.e.:

\[
mG_c - G = \frac{\Gamma}{\sigma^* \rho^*}
\]

where \( \sigma^* \) is the stability parameter, \( \approx 0.025 \), and \( \Gamma \) the Gibbs–Thompson coefficient, \( \approx 0.12 \) K μm (Table 1). For the enlarged dendrite tip in the array, its tip composition
\(C_i = 8.03 \text{ wt}\% \text{ Cu (Fig. 7(b)), which gives } mG_c - G = 31.532. \text{ Thus:}

\[
\rho = \left( \frac{\Gamma}{(mG_c - G)\sigma} \right)^3 = 12.3 \mu m
\]

From the enlarged dendrite tip in Fig. 6(c), seven sets of measurements were made for the width of the dendrite immediately behind the tip. Assuming a parabolic shape of the tip region, the average tip radius measured was 13.2 ± 0.5 \(\mu m\), which is very close to the calculated value from the tip composition measurement. This means that dendrite tip radius is still governed by the local conditions despite the presence of weak laminar convection ahead of the interface in the Al–Cu system.

The above results show that for a weakly laminar flow caused by radial gradients, the morphologies are governed by local values of constitutional supercooling. This observation then allows one to study the microstructural characteristics as a function of interface composition in a given experiment.

4.2.2. Macroscopic interface shape

Three factors are believed to contribute to the formation of a convex interface shape: (1) contact angle restriction at the liquid/solid/ampoule triple junction. This has been observed directly in the transparent systems. In the present study, lack of the critical data for the interface energy between solid Al–Cu alloy and the alumina ampoule hinders further analyses. However, from Fig. 8, it is clear that only a very small part of the interface close to the periphery deviates from the calculated interface shape based on the thermal profile, indicating that the effect exerted by the triple junction is only local near to the ampoule wall. (2) Radial temperature distribution effect. This has been discussed in the numerical modeling part and shown in Fig. 8. (3) The variation in interface composition along the interface causes the interface to become curved to satisfy the local equilibrium condition. By comparing Fig. 2(a) and Fig. 8, it is very clear that a growing planar L/S interface is more convex than a static one. The only difference between them is that a moving interface always needs to maintain a flux balance at the interface, i.e. solute needs to be rejected continually; but this is not the case for a static interface. This solute rejection effect coupled with the flow driven by the radial temperature gradient strongly affects the interface shape, and is considered the third factor responsible for the interface shape. As has been discussed above, a small radial temperature difference is responsible for the slightly convex L/S interface in a static experiment (Fig. 8), therefore the significantly non-flat interface in Fig. 2(a) is due mainly to the radial solute segregation effect. Obviously the latter effect dominates the interface convexity in a growth process.

The first two factors, triple junction constraint and static radial temperature effect, contribute to both the static process (Fig. 8) and the dynamically growing process (Fig. 2). By comparison, their effects on the interface shape are much weaker than the third factor. However, they are the triggering elements for subsequent evolution of the L/S interface shape because the slightly convex interface caused by them leads to a radial solute redistribution in the liquid during growth. The accumulation of solute at the periphery makes the interface lower, i.e. the L/S interface becomes more deflected from a flat interface with the progress of solidification. The same effect has been reported by Kim and Brown [7] for the concave interface evolution in HgCdTe materials in which the solid has a lower thermal conductivity than the liquid.

In conclusion, respective contributions from the three factors, which cause the interface to become convex, can be arranged as:

\[\delta_{\text{triple junction}} < \delta_{\text{radial temperature difference}} \ll \delta_{\text{convection-induced radial segregation}}\]

However, the contact angle at the triple point, i.e. ampoule, solid and liquid junction, can influence the morphology in very thin samples. de Cheveigne et al. [17] and Trivedi et al. [18] have shown that the triple junction constraint may dominate the interface morphological evolution, and they have observed a less stable planar front in very thin samples than predicted by the linear stability theory. Caroli et al. [19] have developed a model that considers the contact angle effect at the triple point junction. They have shown that the contact angle causes the interface to become curved in the transverse direction, and reduces the critical growth rate for a planar interface instability, but only when the thickness of the sample is less than 150 \(\mu m\). In the present investigations, the thinnest sample is of a diameter of 0.6 mm, so that the contact angle effect will be negligible. Thus, the curvature of the interface, observed in the present experiment, is mainly due to convection effects.

4.2.3. Primary arm spacing

Experimental results showed a systematic variation in primary spacing with radial distance. Primary arm spacing is generally correlated with the alloy composition \(C_0\) through the relationship of the form: \(\lambda_1 = K C_0^n\), where \(K\) is a function of growth velocity, temperature gradient and the thermodynamic properties of the alloy and \(n\) is an exponent. Earlier models predict that primary arm spacing increases with alloy composition, and this was verified experimentally by Trivedi and Somboonsuk [20].

A detailed numerical model of primary spacing is given by Hunt and Lu [3]. They have given analytical expressions for the smallest stable primary spacing. This model cannot be applied directly to analyze the present experimental results since in the experiments the bulk composition is fixed but the composition and the composition gradient in the liquid at the cell or dendrite tip is altered due to the
Fig. 10. Calculated primary spacing range based on Hunt–Lu model [3] for the cells and dendrites with \( G = 6.5 \text{ K mm}^{-1} \) and \( V = 7.65 \text{ \mu m s}^{-1} \). The experiments fall within the range for cell spacing.

presence of fluid flow. Thus, the primary spacing needs to be correlated with the tip composition rather than with the bulk composition. In order to obtain a general trend in the spacing variation, the following procedure was adopted. Since both the tip composition and the primary spacing results are given as a function of bulk composition, \( C_0 \), the value of \( C_0 \) that gives the measured value of tip composition, \( C_r \), was first calculated. Then this value was used as the bulk composition to calculate the lower limit of primary spacing. The upper limit of primary spacing is predicted by Hunt and Lu [3] to be about twice the minimum spacing value. The results of the predicted spacing and experimental spacing are shown in Fig. 10. Note that the measured values of cellular spacing agree remarkably well with the average spacing predicted by the model. Note also that in the region where cells are formed, the primary spacing variation with distance is negligible. This is expected, since the cell tip composition variation, shown in Fig. 6(b), is negligible.

The value for the dendrite spacing is larger than the cell spacing, but much smaller than the value predicted theoretically. This is expected, since the dendrites observed in this study are not well developed with respect to side branch formation on both sides. In fact, the branching is seen to occur only on one side and no branches are observed in the direction towards the cell. Thus, the dendrite that has been observed experimentally is not a fully developed dendrite that is surrounded by similar dendrites, so that the observed spacing, although higher than the cell spacing, is still significantly smaller than the predicted steady-state dendrite spacing. It should be noted that whilst the primary spacing is smaller, the tip radius value matches well with the theoretical value. This is of course understandable, since the dendrite tip radius value adjusts rapidly, whereas the primary spacing dynamics are much slower and take a long time to reach the steady-state value, as shown by Somboonsuk and Trivedi [21] and Han and Trivedi [22].

5. Summary

The effect of convection on microstructure development in Al–4.0 wt% Cu alloys has been examined theoretically. A numerical model is developed that takes into account the radial temperature gradient that is generally present in Bridgmen growth. The effect of convection is examined as a function of sample diameter, and it is shown that a sample diameter of 1.0 mm or less is needed to obtain a diffusive growth in the Al–Cu system.

A new experimental technique is used in which several samples of small diameters are placed inside a large ampoule, and directionally solidification simultaneously in the same experiment. Since the convection effects become smaller in finer diameter samples, the variation in microstructure with sample diameter can be correlated with the extent of convection. The development of interface shape and microstructure scales are then examined as a function of sample diameter, and it is shown that the non-planarity of the interface increased as the sample diameter was increased. Different phenomena that lead to the curvature of the interface have been discussed, and it has been shown that the solute redistribution is the dominant effect.

In large diameter samples, convection causes the interface composition to increase from the center to the periphery, giving rise to non-uniform microstructure formation. Through critical experiments near to the morphological transition conditions, a transition in morphology from planar to cellular or from cellular to dendritic is examined in a given sample. Detailed measurements of interface composition have been carried out, and the results
analyzed to show that the location of the transition could be related to the local conditions of interface composition and thermal gradient. In addition, microstructure scales, such as cellular spacing and dendrite tip radius, have been shown to follow theoretical predictions if local measured values of the interface composition and temperature gradient are used in the model. These observations allow one to analyze quantitatively the seemingly complex microstructures that form under convective growth conditions, and provide valuable information on the effect of interface composition on morphological development.

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