

The Mechanisms of Formation and Prevention of Channel Segregation during Alloy Solidification

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Conditions for the formation of macroscopic segregation channels have been examined in the ammonium chloride-water and lead-tin systems, using base chilled molds. Such channels develop when the rejected solute is less dense than the solvent and are therefore a result of density inversion, but slow (<5 rpm) rates of mold rotation, about axes inclined to the vertical by 20 deg to 30 deg, throughout the time of solidification, effectively prevent the formation or propagation of these channels. Artificially created channels or those momentarily blocked fail to continue and are overgrown, but channels can be initiated by drawing liquid upward from close to the growth front in fine capillaries. Examination of these effects leads to the conclusion that channels originate at the growth front, rather than within the dendritic array, and that their formation is necessarily preceded by a liquid perturbation from the less dense boundary layer into the supernatant, quiescent bulk liquid. Intermittent 'solute fingers' are then fed by dendritic entrainment to produce stable convective plumes and concomitant channels. It is considered that the effects of mold precession are primarily caused by translation of bulk liquid across the dendritic growth front, shearing off convective perturbations from the boundary layer before they have time to develop. The nature of the liquid movements is discussed and shown to be a function of the mold dimensions. The inclination of the gravitational vector within the solid-liquid, dendritic array is considered to be of secondary importance to the formation or prevention of channels.

I. INTRODUCTION

DURING alloy solidification through a significant freezing range (e.g., ≈ 50 K) over times in excess of $\sim 10^3$ s, it is frequently found that nearly vertical, solute-rich channels develop within the dendritic mesh of the partly solidified ingot: these are typically $\approx \times 5$ wider and some two/three orders of magnitude longer than the interdendritic spacings. Such structural defects are highly deleterious for the mechanical properties of a product. In cast billets, such channels are termed 'A' segregates and run approximately at right angles to the horizontal heat flow direction; in directionally solidified ingots with predominately base chill, they run antiparallel to the heat flow direction and produce so-called 'freckles' on outer surfaces or on horizontal sections.

It has been clearly demonstrated that channel development occurs in alloys where the solute is less dense than the solvent, so that interdendritic and boundary layer liquid is less dense than bulk liquid. Channel activity has been illustrated in the transparent/luculent $\text{NH}_4\text{Cl}-\text{H}_2\text{O}$ system¹ with side chill^{2,3} and base chill⁴ configurations, and in the former case it has further been demonstrated that ternary additions which reverse the solutal density gradient also reverse the direction of interdendritic flow.^{3,5} Changes in bulk liquid composition are also related to similar density inversion caused by boundary layer⁶ and/or by minor thermal variations in radial temperature gradients within a cylindrical vessel (e.g., References 7 and 8).

In the two geometrical configurations — with horizontal vs vertical heat flow — the overall patterns of fluid flow have to be somewhat different; thus, in the former there

can be long-range fluid flow patterns (Figure 1(a)), while in the latter, the possible flow pattern must be restricted to relatively local entrainment close to the dendritic growth front (Figure 1(b)). The present paper relates to the second configuration which is the simpler to analyze, although both must be closely related.

Although expressions have been presented for general interdendritic flow under the combined influences of contraction and convection,⁹ there has, as yet, been no attempt to measure or model the formation and dimensions of channel patterns in either geometry.

Further, it has been shown that mold movements which cause the bulk liquid to translate across the growth front can prevent channel formation or inhibit channels previously established in a stationary base chilled configuration. This has been demonstrated in $\text{NH}_4\text{Cl}-\text{H}_2\text{O}$ ⁸ and $\text{Pb}-\text{Sn}$ ¹⁰ alloys. The movement which is effective is slow, ≈ 5 rpm, rotation about axes inclined to the vertical at up to 20 to 30 deg, and as such, centripetal forces are not sufficient to contribute significant radial segregation:¹¹ slow rotation about a vertical axis is not effective. The effect of this type of movement is to cause the gravitational vector to precess about the surface of a cone of semi-apical angle equal to the tilt of the axis of rotation (Figure 2). Originally,⁸ it was suggested that this movement would inhibit or retard channel development within the dendritic mesh by preventing fluid flow in any single direction for significant time. However, as will be evident from what follows, it is now thought that the effect on bulk liquid movement is the more important factor.

Some comparisons between the systems and materials parameters are desirable: Figures 3(a) and (b) show the relevant phase diagrams or portions thereof; Table I lists some relevant parameters.

In the aqueous system, allowing sufficient superheat to prevent general 'equiaxed' grain nucleation,¹ the upper practical pouring temperature is ~ 100 °C or a maximum working concentration of ~ 35 wt pct NH_4Cl . From the phase diagram and densities, the maximum fraction solid,

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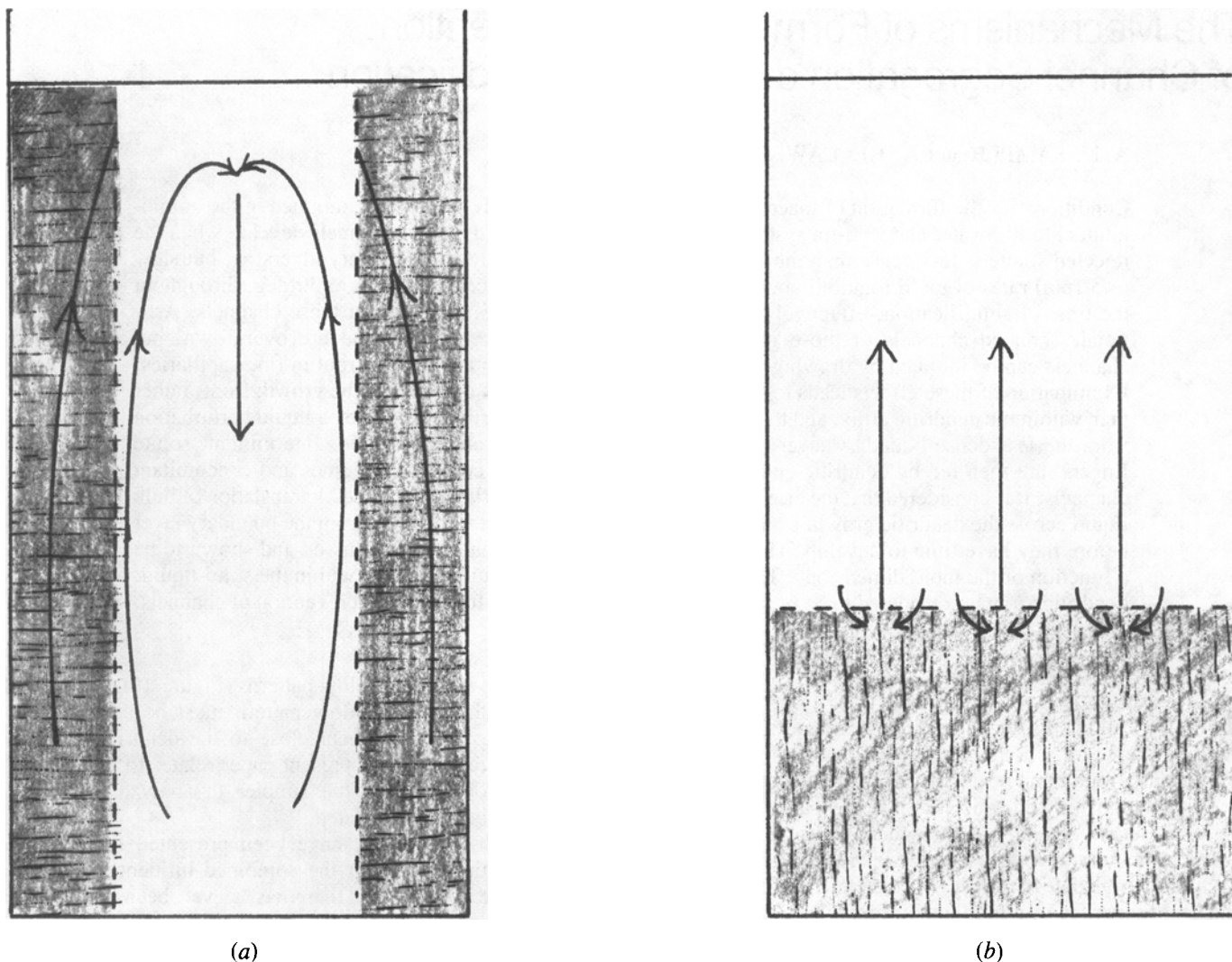


Fig. 1—Indicating schematic fluid flow patterns in partly solidified ingots when the solute is less dense than the solvent: (a) with heat flow horizontal and (b) with heat flow vertically downward.

Table I. Comparative Data for the $\text{NH}_4\text{Cl-H}_2\text{O}$ and Pb-Sn Systems

System Parameter	$\text{NH}_4\text{Cl-H}_2\text{O}$	Pb-Sn
Volume fraction liquid at the eutectic temperature and 35 wt pct NH_4Cl , 10 wt pct Sn	0.86	0.09
Pct change of volume on freezing of primary dendrites	~ -30	-3.8
Pct change of volume on freezing of eutectic mixture	+0.5	-3.0
Liquid densities on liquidus at 35 wt pct NH_4Cl , 10 wt pct Sn (g/cc)	1.08	9.98
ν -kinematic viscosity of liquid on liquidus at 35 wt pct NH_4Cl (centistokes), 10 wt pct Sn	0.95	0.25
Thermal conductivities: $\left(\frac{w}{mK}\right)$		
(a) solid	2.5×10^{-6}	35.0
(b) liquid	0.49	18.0
(c) eutectic mixture	2.70	55.0
Prandtl no. (est)	~ 7	~ 0.02

just above the eutectic temperature at -16°C may then be estimated at ~ 16 pct and the dendritic mesh is therefore very open or permeable to fluid flow. In contrast, a Pb-10 wt pct Sn alloy, solidifying over about twice the temperature range with segregation according to the Scheil equation, would have a fraction solid >90 pct at the eutectic temperature of 183°C and thus be much less permeable.

With reference to the relative densities, it will be observed (Table I) that the contraction on freezing for primary

NH_4Cl is very large (≈ 30 pct) while there is actually a small expansion for the $\text{NH}_4\text{Cl-H}_2\text{O}$ eutectic solidification. By comparison the primary lead solid solution undergoes a much smaller contraction on freezing (≈ 3 pct), and there is a small contraction on passing through the eutectic. However, combined with the relative volume fractions at the eutectic temperatures the primary and total contractions on freezing are not dissimilar, *i.e.*, ~ 4.8 pct vs 3.5 pct and 4.4 pct vs 3.7 pct, respectively, in each system.

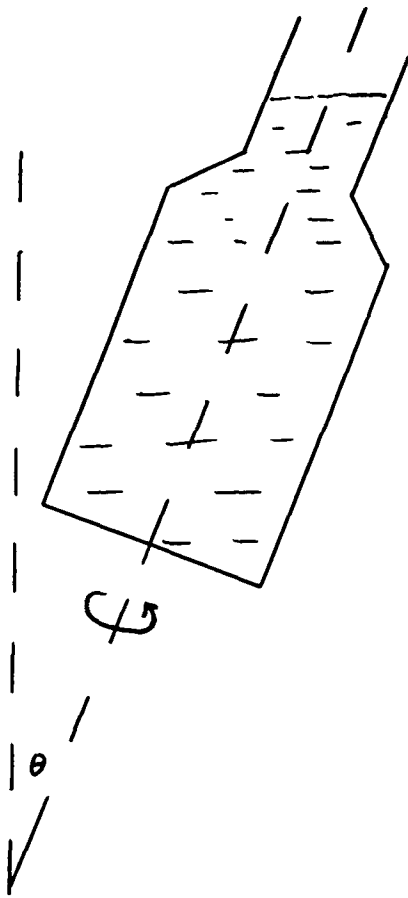


Fig. 2—Type of rotational or precessional movement employed.

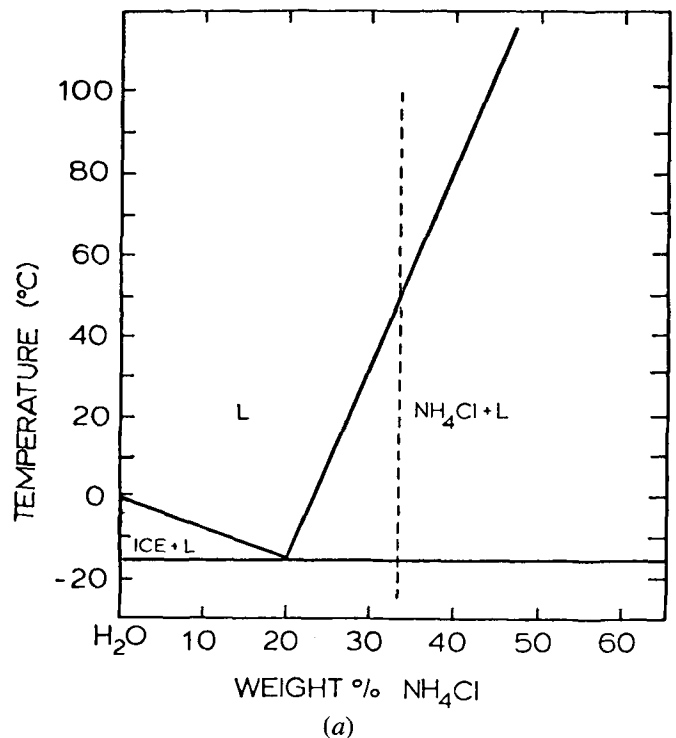
Comparison of the dynamic and kinematic viscosity coefficients also shows relatively small differences, the latter being the more important in that it appears in the Reynolds number ($\nu r/\nu$), and with measured flow rates, ν , in the aqueous system and mean channel radii, R_c , in either system (see later), the Reynolds numbers are thought not to exceed 10^2 , which is well within the range for streamline flow ($\lesssim 10^3$), despite the fact that the channels or interdendritic capillaries are irregular in section.

Otherwise, the major difference between the two systems is probably in the thermal conductivities (Table I), and the much lower values for the nonmetallic phases mean that the overall temperature gradients through the growth front will be much steeper in the aqueous system and that local gradients around perturbations will also be correspondingly larger as heat is conducted away less rapidly; this thermal inertia is indicated by a much larger Prandtl number (ν/D_{thermal}) than for the metallic system, implying that as far as heat flow is concerned, perturbations will be less easily damped.

II. EXPERIMENTAL

A. $\text{NH}_4\text{Cl}-\text{H}_2\text{O}$ System

The base chilled arrangement⁸ was essentially that described by Copley *et al.*⁴ and consisted of a 75 mm diameter plexiglas cylinder, height 160 mm, sealed by an 'O' ring onto a copper base which was cooled by a reservoir of liquid nitrogen at -196°C . Salt solutions were poured into this



Pb-Sn Lead-Tin

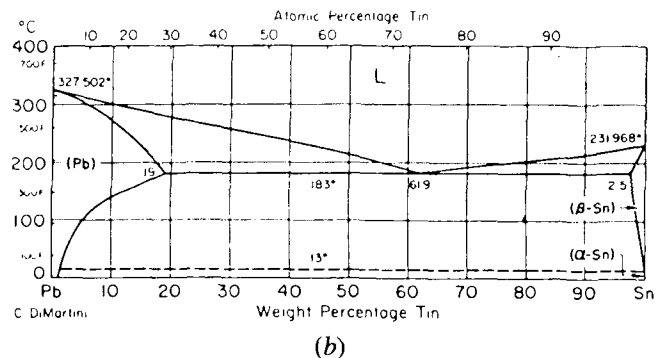
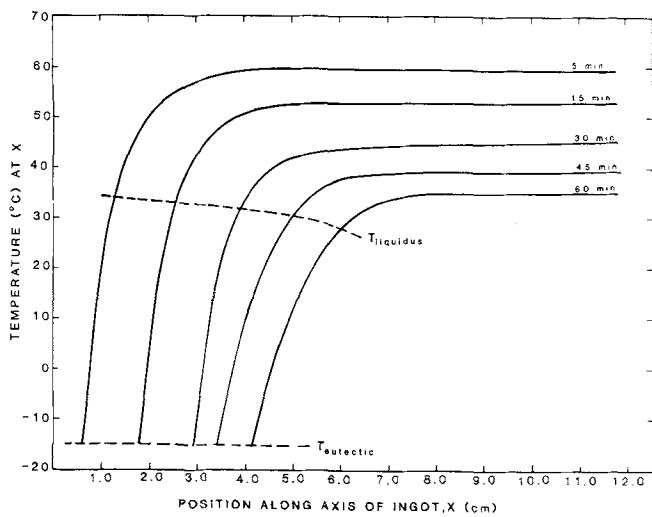


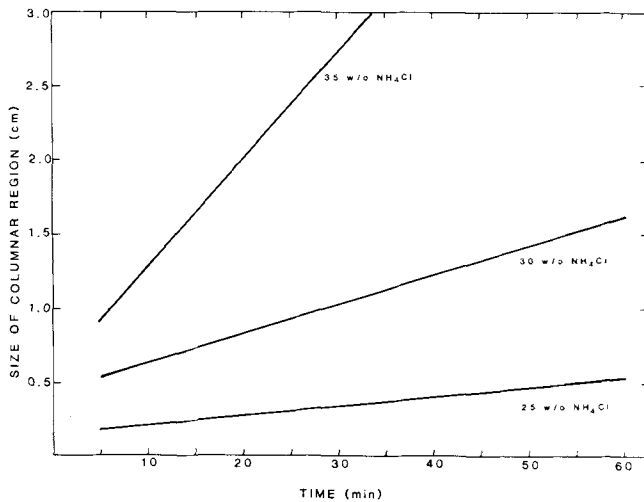
Fig. 3—Partial phase diagrams for the systems $\text{NH}_4\text{Cl}-\text{H}_2\text{O}$ and Pb-Sn.

mold with a constant superheat of 35 K above the liquidus, and subsequent events then recorded photographically from the side and above. Salt solutions containing 25 wt pct, 30 wt pct, 35 wt pct NH_4Cl were used; these would have liquid fractions at the eutectic temperature of ~ 95 pct, ~ 90 pct, and ~ 84 pct, respectively.

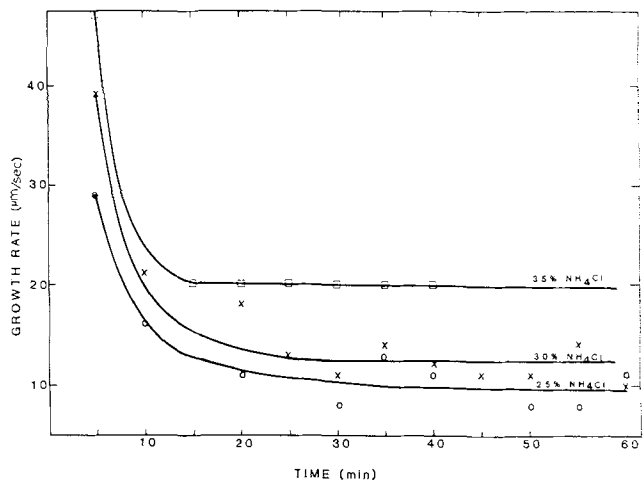
As the solution freezes upward from the chilled base it becomes resolved into three zones: (a) supernatant bulk liquid, (b) primary dendrites with interdendritic liquid, and (c) primary dendrites in the solid eutectic mixture. The extents of these zones were measured on the sides of the mold and correlated well with vertical temperature profiles which were recorded by a fine chromel-alumel thermocouple enclosed in an axial, thin walled, glass capillary. These combined measurements then appear as in Figures 4(a) through (c) as temperature profiles, extents of middle zone, and growth rates of the dendritic front. As expected, the extent of the dendrite-liquid zone increases with concentration and almost linearly with time (b), while the growth rates decrease rapidly to nearly constant values (c).



(a)



(b)



(c)

Fig. 4—Showing (a) vertical temperature profiles along a 30 wt pct NH_4Cl casting at various times—positions of dendritic and eutectic fronts as indicated, (b) heights of dendritic zone vs time for three compositions, and (c) the growth rates as functions of time and composition.

Movement of the mold was carried out by mounting the assembly upon a board which could be tilted and rotated about an axis which coincided with that of the cylindrical mold.

B. Pb-Sn System

A Pb-10 wt pct Sn alloy was used; this was melted and poured into a cylindrical graphite crucible of i.d. 40 mm and height 120 mm, at a temperature of 600°C (i.e., with 300 K superheat). The crucible was placed in a resistance furnace at 550°C with the base of the crucible resting on a water cooled support at the bottom of the furnace, and the furnace was then allowed to cool at a rate determined by a programmed Eurotherm controller, the temperature at the top of the melt being recorded down to the eutectic temperature, a rate of ~ 0.75 K per minute allowing for a period of several hours for complete solidification.

To examine the effects of mold rotation during solidification, the entire furnace assembly was mounted on a circular base plate with axial pins below of various lengths to determine the angle of inclination (Figure 5). The frame containing the assembly could then be rotated by an arm above, driven by a geared motor. By allowing the edges of the base plate to slide on a teflon sheet and with light spring loading, the furnace precessed smoothly about the axial support pin without rotation about its own axis, thereby avoiding complications with electrical and water supplies. The assembly could be precessed in this way about axes inclined from ~ 5 deg to 25 deg to the vertical, at up to 10 rpm.

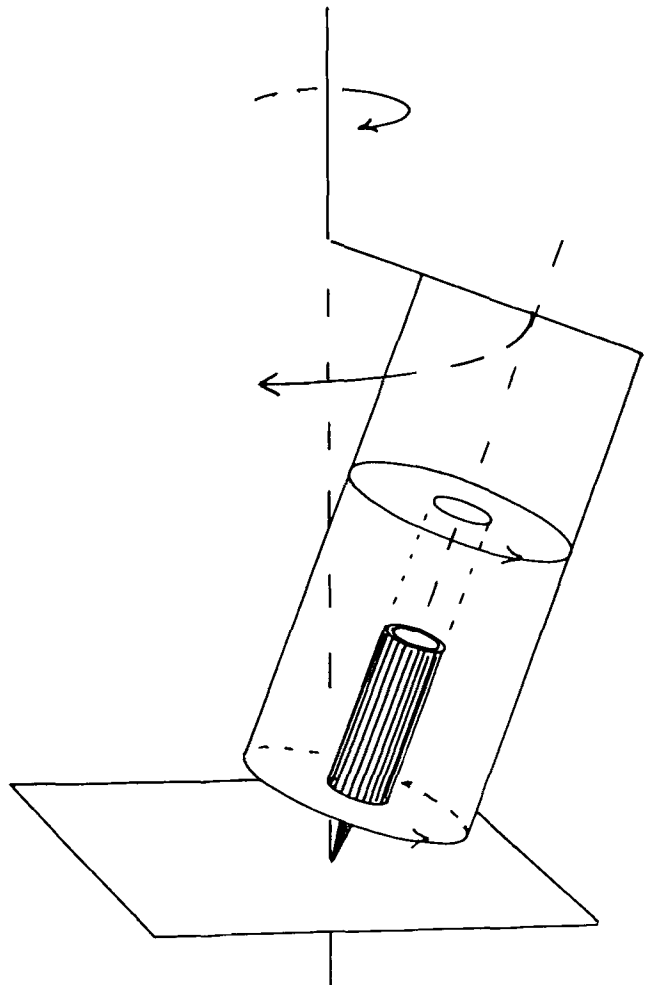


Fig. 5—Schematic diagram of assembly used to solidify and precess Pb-Sn alloy.

III. RESULTS

A. $\text{NH}_4\text{Cl-H}_2\text{O}$ System

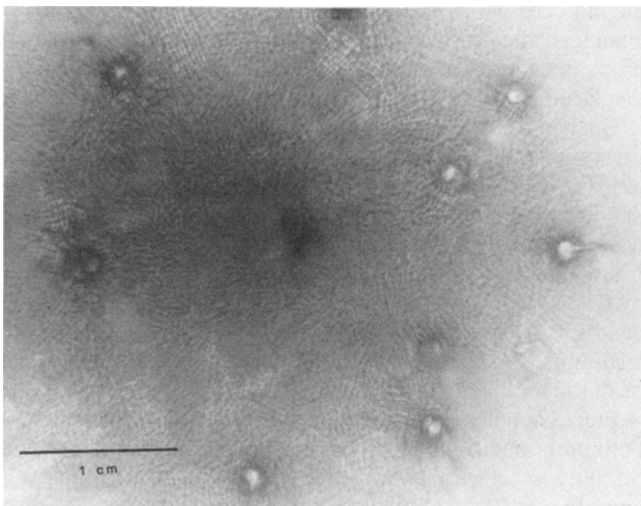
1. Channel formation

With the mold stationary, vertical, or inclined, or rotating at <5 rpm about vertical or inclined axes, the macroscopic growth fronts were parallel to the chilled base, *i.e.*, at right angles to the axis. At rates of rotation >5 rpm and up to 10 rpm there was increasing evidence that the less dense aqueous solution was being centripetally driven toward the centerline, so that the freezing point was depressed and a shallow sump developed, *i.e.*, the interface became macroscopically concave toward the liquid.

In the 25 wt pct NH_4Cl castings no discernible channels were observed over 1 hour, after which growth effectively ceased. As noted above, the extent of the mushy zone was never greater than ~ 5 mm (Figure 4(b)), the maximum fraction solid did not exceed ~ 5 pct, and the primary dendrite array was very open and permeable.

In the 30 wt pct alloy very fine channels developed after some 5 to 10 minutes, while in the more concentrated solutions the incubation times were longer, 15 to 20 minutes, but channels were then larger and well defined (Figure 6(a)), and were each associated with a plume of less dense liquid (Figure 6(b)) rising steadily to the upper surface, a height of about 100 mm, as reported previously.^{4,8,10}

During the incubation period, *e.g.*, ≈ 10 minutes, there was evidence of many fine fluctuating and decaying solute plumes rising from a boundary layer at the dendritic front (Figure 7). These were initially concentrated toward the center of the mold and contributed to a general slow convective mixing of the bulk liquid upward in the middle and downward at the sides. This pattern was attributed to small radial temperature gradients, and it is known that even with extreme care⁷ this thermal convection is very difficult to eliminate. As a consequence of this convective mixing, the growth of the front is not quite steady state and the bulk liquid becomes less concentrated—this has been confirmed by chemical analysis⁸ and can be seen in Figure 4(a) where the level of the dendritic front gradually falls to lower temperatures at longer times.



(a)

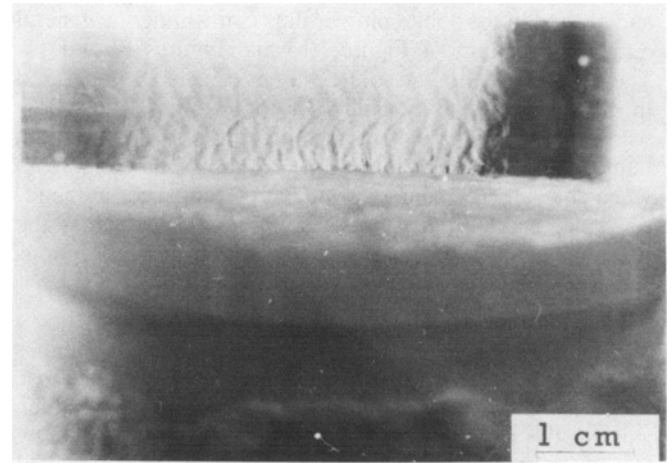
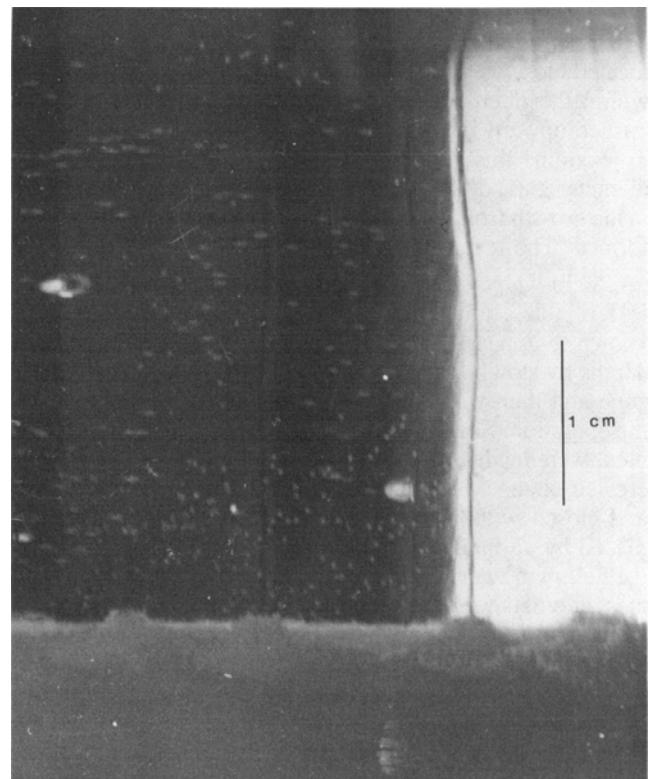


Fig. 7—Small turbulent perturbations arising from boundary layer ahead of dendritic growth front, prior to established channel development.

Established channel dimensions are up to $\times 4$ the mean spacings of the square cross grid of the NH_4Cl dendrites, and the mean spacings of channels are about 10 mm or ~ 25 interdendritic spacings. Between the 35 wt pct solution and 32 wt pct solution, used in previous work,⁸ it was not possible to detect any significant differences in dimensions or incubation times.

The solute plumes and flow patterns can be better identified with a soluble coloring. Small potassium permanganate crystals were placed on the growth front approximately midway between established channels—subsequently, within ~ 1 minute, colored liquid was entrained into the channels



(b)

Fig. 6—(a) Top view of channel pattern in 30 wt pct NH_4Cl casting and (b) solute plume rising from one such channel; side view.

and ejected in the solute plume, thus confirming the general circulation pattern of Figure 1(b) and Figures 10 and 11. The flow rate within the solute plumes can also be measured in this way and was typically 7.5 to 10 mm s^{-1} ; this flow was visibly streamlined, as is consistent with a Reynold's number of $\lesssim 10$ for plumes of $\sim 1 \text{ mm}$ diameter. Even when channels and plumes were well established, a boundary layer was still discernible as a faint film across the dendritic front, from which minor unsuccessful eruptions occurred periodically.

2. Mold movements

With a stationary but inclined mold, channels and plumes developed in about the same time as previously and then rose vertically so that the channels were inclined to the specimen axis by the angle of tilt—this is as described earlier by Copley *et al.*,⁴ both for this aqueous system and in a tilted E.S.R. steel ingot. Slow rotation (*i.e.*, $\lesssim 5 \text{ rpm}$) about a vertical axis did not change the pattern of channel development from that described in vertical stationary ingots.

With mold rotation about axes inclined between 20 deg to 30 deg , at rates rising from 1 to 5 rpm , channel development was effectively inhibited throughout a one-hour period. Moreover, if channels were allowed to develop with rotation about a vertical axis and the mold then tilted to rotate about a 30 deg axis, channels closed within two/three minutes, reforming elsewhere if the mold was subsequently returned to the vertical. The noticeable and obvious difference between vertical and inclined rotation is that in the latter case the bulk liquid is continuously translated in a circular shearing movement across the growth front while in the former there is little such translation at these slow rotation rates.

3. Disturbance of the growth front

It is inferred from the preceding that channel activity is located close to the growth front, where liquid is entrained within the interdendritic capillaries between channels and ejected upward from the channel core into the bulk liquid. To examine this further, trials were made in which 1 mm diameter glass quills were used to puncture holes artificially in the growth front and also to block those which had already formed. The results were informative:

(a) A $30 \text{ wt pct NH}_4\text{Cl}$ casting was allowed to grow for $\gtrsim 15$ minutes, after which time well-defined channels are known to appear. The front was then punctured to various depths by slowly punching the pointed quill into the mushy zone and then withdrawing it, a period of ~ 5 seconds for the operation. Single and multiple holes, "typically" distributed, were made up to depths of $\sim 10 \text{ mm}$. These artificially created channels failed to propagate and were overgrown and closed within 2 to 4 minutes, subsequently being replaced by an independent array.

(b) Following normal channel formation at times > 20 minutes, attempts were made to even the channel distribution by inserting the quill at relatively open parts of the front where spacings were large—these also failed to develop.

(c) If an established channel was blocked by a quill for 5 to 10 seconds, it subsequently closed within a few minutes, sometimes being replaced by another independent channel in the same area.

(d) Some comments may be added, here, about the adjustment of channel spacings. It may be seen in Figures 6(a) and 8(a) that some well-defined channels are closer together than

the average spacing; such adjacent channels do not appear to diverge or combine in any very clear manner and may continue to grow nearby for extended periods (*e.g.*, 10 minutes). The channel arrangement, when it changes, does so by one or both channels closing off, sometimes being simultaneously or subsequently replaced by a third.

(e) The above experiments were repeated with the same results but using an open quill which had been sharpened at the rim edge and could be twisted on its axis when inserted into the growth front. This insert then cuts its way into the dendrites like a fine drill—the reason for this alternative was to ensure that the previous results were not a consequence of the dendrite mesh being locally crushed, causing reduced fluid flow by blocking entrainment.

4. Disturbance of the bulk liquid

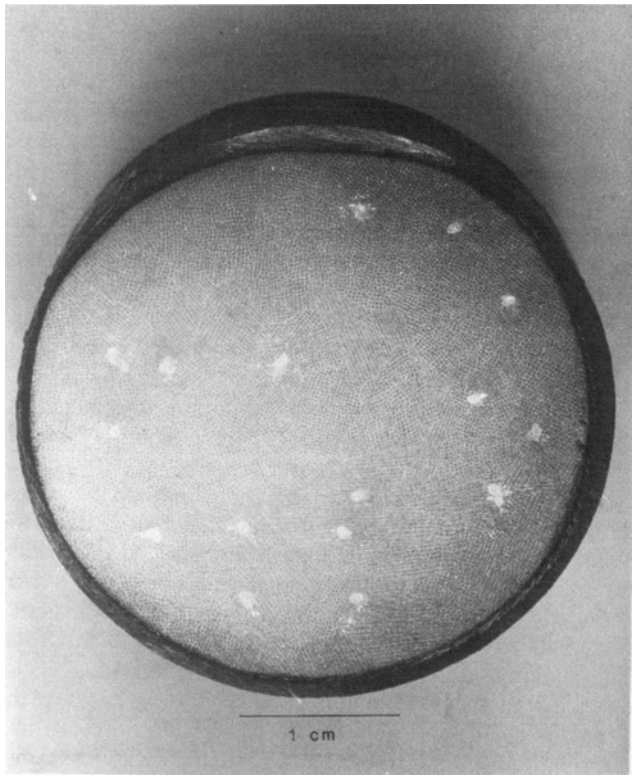
Since—from the preceding trials—it seemed impossible to initiate channels artificially within the mushy zone, the alternative experiment was made by locally disturbing the bulk liquid close to the growth front. To do this, an open capillary was slowly lowered down to within 1 to 2 mm of the growth front and liquid was then drawn up the tube to create an artificial solute plume from the boundary layer. This flow was maintained for a $\lesssim 10$ seconds and the capillary was then withdrawn steadily out of the bulk liquid; the operation was rather delicate and care was taken not to agitate the bulk liquid in any other way. In the course of five trials, five successfully 'nucleated' channels appeared at the position where a plume had been created.

B. Pb-Sn System

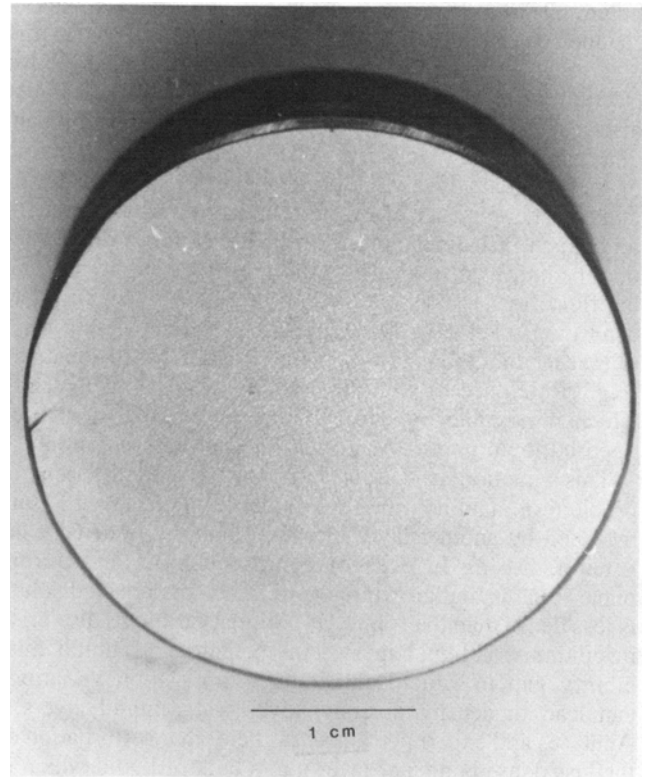
Channel Formation and Prevention: The cooling conditions described previously gave well-defined channels in at least the upper two-thirds of ingots. Figure 8(a) is an example of the macroscopic pattern, and Figure 8(b) shows the microstructure of one such channel. The channels were visible as tin-rich material, with an irregular two-phase structure, presumably of approximately eutectic composition. The square grid of the dendritic array and columnar grain structure are clearly visible. The general pattern of channels could be followed through on successive sections and did not change dramatically. If the assembly was tilted, channels also formed, as in the aqueous analogue, and propagated vertically, as could be followed by the lateral displacement on successive transverse sections.

Quenching experiments and examination of vertical sections showed that the macroscopic growth front was planar, as in the analogue castings. It was particularly important that the furnace temperature, position of mold, and efficiency of water cooling be correctly adjusted to maintain such a front—and this had to be done by trial and error. As noticed by Copley *et al.*,⁴ failure to obtain an approximately planar dendritic front resulted in channels being restricted to the leading part of the front, *e.g.*, in the middle or at the sides of the ingot.

Precessional movement of such ingots showed that the optimum conditions for channel prevention in a 10 wt pct Sn alloy were with 1 rpm rotation about a 20 deg inclined axis (Figure 8(c)). A rate of 5 rpm appeared to be too fast and caused some evidence of Sn concentration toward the center of ingots from centripetal effects. An axial inclination of only 10 deg proved insufficient for complete channel



(a)



(c)



(b)

Fig. 8—Lead-10 wt pct Sn ingot (a) with established channel pattern, (b) showing channel detail, and (c) structure produced under same conditions but with assembly rotated at 1 rpm about an axis inclined at 20 deg to the vertical.

lower, *i.e.*, 1 rpm vs 5 rpm, and the mold was narrower, *i.e.*, 40 mm diameter compared to 75 mm in the aqueous experiments.

IV. DISCUSSION

From the preceding it is necessary to consider (a) channel nucleation, (b) channel propagation, and (c) the effects of mold movements. With the data presently available most of the information about (a) and (b) comes from the transparent aqueous system, but the metallic system offers the possibility of varying the extent and permeability of the 'mushy' zone over a much wider range.

(a) Channel Nucleation

'Nucleation' is perhaps not the correct term, but there is a clear incubation period before channels begin to appear and this, in a vertical mold (stationary or otherwise), corresponds approximately to a time and position at which the dendritic growth rate has declined to an approximately constant level. Experiments with interrupted mold rotation/inclination show that channels renucleate fairly rapidly. In this connection, however, the experiments with artificially created or blocked channels are particularly instructive, and it would appear that for a channel to develop there must be a simultaneous solute plume in the nearly quiescent bulk

elimination. Compared with the aqueous system, the conditions for channel prevention must be considered remarkably similar, but the necessary rate of rotation was significantly

liquid. Clearly, there can be no steady state plumes without channels to supply the flow, but it would seem that the reverse is also true, and the experiments described above (section III-A-4) show fairly conclusively that bulk liquid disturbance is a necessary precondition for the development of a channel. The suggestion made here is that a perturbation of the boundary layer is a prerequisite for channel development, the sequence of Figure 9 indicating what is thought to happen. Channels must originate at the front rather than behind it in the bulk of the mushy zone, because all the fluid flow is continuous and there is no way an interdendritic pocket at some depth can accelerate without prior release at the growth front. The parallel has been made (*e.g.*, Reference 8) with the draining of a stagnant swamp which is possible only by creating an external breach as opposed to an internally created channel with no outlet.

This situation is therefore connected with the general problem of stability when a less dense liquid layer is surmounted by another layer in which there is no density inversion, arising in oceanographic contexts¹² and during plane front crystal growth upward, when the rejected solute is less dense than the solute.¹³⁻¹⁶ Similar problems also arise in columns of liquid in systems containing a liquid miscibility gap in which temperature-composition variations can lead to density inversion over wider liquid layers.¹⁷ Analyses and experiment show (as here) that perturbation of the less dense boundary layer into a bulk of liquid which is situated in a positive temperature gradient is not necessarily

spontaneous, when the positive density gradient arising from composition gradient exceeds the negative gradient caused by the coefficient of thermal expansion.

In the case of a planar interface covered by a narrow boundary layer, analysis shows that there can be a significant range of growth rate and composition, for a given temperature gradient, where the boundary layer is unstable to perturbation although the plane front remains stable to breakdown.^{14,15} Whereas the critical concentration for interfacial breakdown decreases with increasing growth rate, that for liquid perturbation actually increases.⁷ A physical interpretation of why this should be so might be that an increase in growth rate leads to a narrower boundary layer (D/V), within which the viscous resistance to lateral flow, necessary to create a perturbation—*e.g.*, Figure 9—increases, therefore requiring a bigger driving force from the solutal-density inversion.

It is concluded from the experiments described above that the situation is qualitatively similar ahead of a dendritic front, but there are some obvious differences. The boundary layer is of the same order of thickness, characterized by D/V , and within that layer the possibility of convective perturbations must be similar to that ahead of a plane front. The situation differs, however, in that fluid flow is not restricted to that within the boundary layer because the 'solid' is itself permeable, and a perturbation upward into the bulk liquid can be more readily supplied with less dense liquid. Thus, a sequence such as that of Figure 9 would

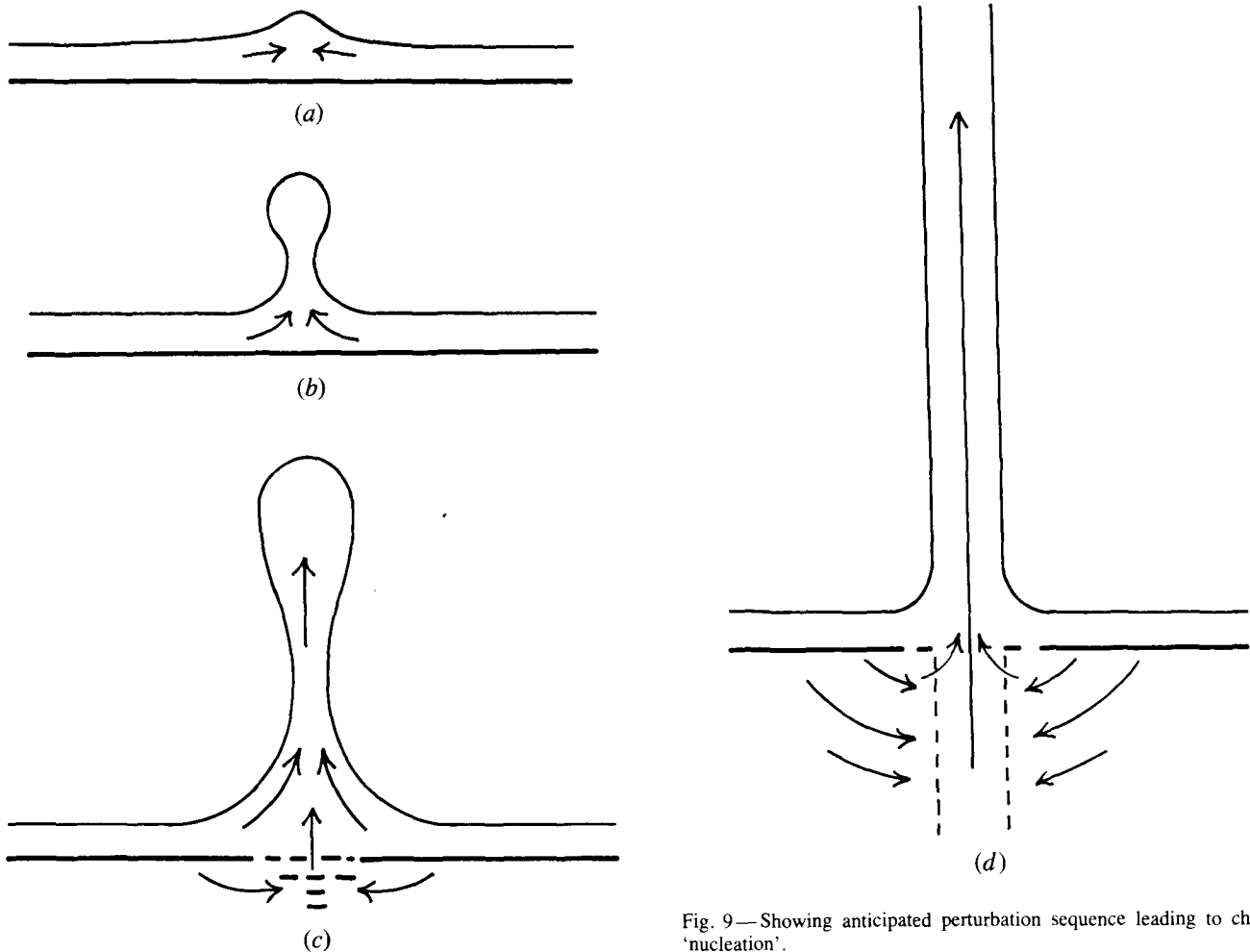


Fig. 9—Showing anticipated perturbation sequence leading to channel 'nucleation'.

seem to be the most probable, entrainment through the dendritic mesh leading to a continuous and near steady-state cycle with long solute plumes such as cannot be maintained ahead of a plane front, nor, probably, a cellular front.

Of course, a mathematical analysis can relate only to a plane interface so that convective perturbations occur everywhere with equal probability and some characteristic wave length.^{15,17} A dendritic front is less perfect, there being minor variations in dendritic spacing within any one grain and larger irregularities at grain boundaries and triple junctions—undoubtedly these must tend to localize the perturbations which lead to channels. However, examination of sections such as that of Figure 8(a) does not reveal any preferential location of channels at grain boundaries, but this may not be significant because both channels and boundaries may have diverged during growth from the original events.

(b) Channel Growth

Here, we are concerned with continuous entrainment, downward into the dendritic front between channels, laterally and radially inward to the channels and upward in the plume toward the upper surface of the bulk liquid (a distance $H \approx 100$ mm).

Within a plume, flow is visibly streamlined, and from Poiseuille's equation the volume transported upward per unit time, V , is given by

$$V = \frac{\pi \Delta P R_c^4}{8H\eta}$$

where ΔP is the pressure drop along the plume, R_c = the plume radius, and η the viscosity (~ 1 Poise), indicating a pressure of ≈ 10 N m⁻² from observed flow rates.

Around any one channel the entrainment volume can be described with reference (Figure 10) to a cylinder of radius R_0 and depth h_0 , below which it is supposed that sufficient liquid has been drawn into the channel to feed the plume, so that liquid farther down is essentially stagnant. Around the circumference at R_0 the liquid would also be idle, unless adjacent entrainment volumes overlapped. There does not, from observation, appear to be a very regular channel spacing, *e.g.*, Figures 6(a) and 8(a), so that competition between channels does not obviously determine the spacings very

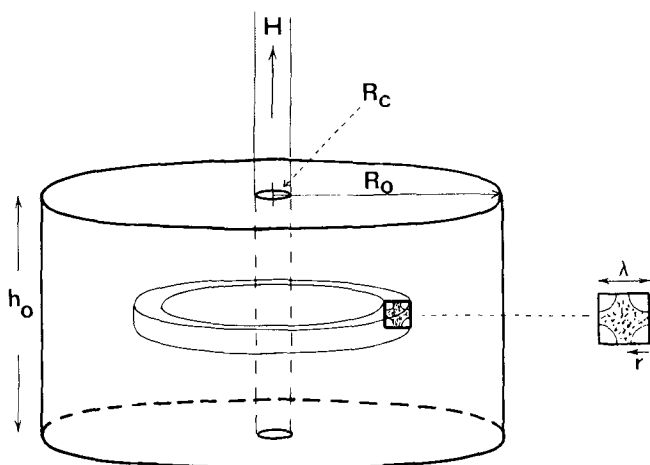


Fig. 10—Showing reference volume element for entrainment of liquid into channel and plume.

rigorously. It might be inferred from this that channel spacing is primarily determined by the chance positions at which they nucleated.

Within the reference volume the interdendritic capillary flow is presumably most rapid close to the channel mouth, where a rim or cone develops, and is therefore of semi-ellipsoidal form, as in the contours of Figure 11. Flow in the interdendritic capillaries is estimated from the experiments with dye, to occur with a mean velocity, v , of about 0.1 mm s⁻¹, and although the capillaries are of periodically fluctuating radius, r , the flow would probably also be streamlined on a basis of an estimated Reynolds' number of < 1 .

With a square array of dendrites of spacing λ , the number of capillaries per unit cross sectional area is

$$N = \frac{1}{\lambda^2}$$

and the fraction of liquid per unit area

$$f_{L/A} = \frac{\pi r^2}{\lambda^2} = f_{LV}^{2/3}$$

where A and V , respectively, denote area and volume.

Substituting these quantities again into a Poiseuille form gives an interdendritic flow per unit time and area

$$v = \frac{\Delta p f_{L/A}^2 \lambda^2}{8\pi \rho \eta}$$

where Δp is now a pressure difference, as between the growth front and channel walls, and is difficult to assess or express satisfactorily.

The $f_{L/A}^2$ is the dimensionless permeability of the dendritic mesh, similar to the quantity, K , used by Mehrabian *et al.*⁹ but without a factor to account for the imperfections of interdendritic capillaries. In their⁹ analysis for general three dimensional flow there was no expression of interdendritic spacings because they were concerned with relative contributions rather than the situation here which is constrained to supply the suction up a channel plume. Thus, the plume

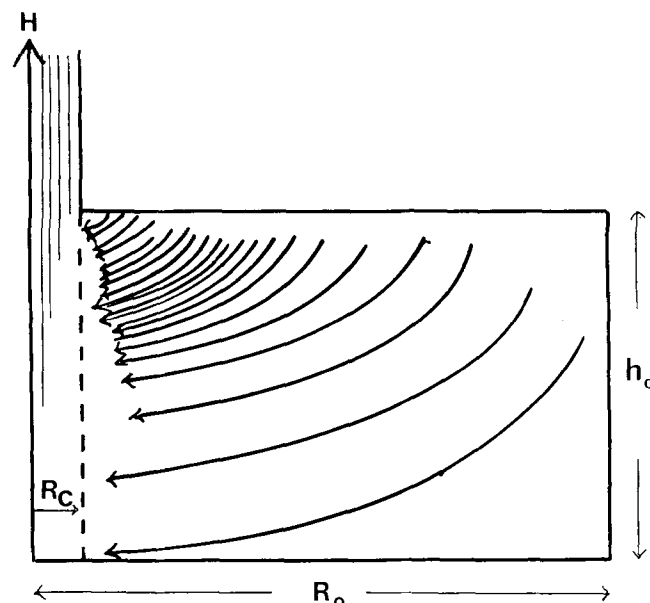


Fig. 11—Probable flow contours to feed plume.

flow, V , must equal the combined capillary flows around channel walls of circumference $2\pi R_0$, summed from $h = 0$ to h_0 at the base of the reference volume.

(c) *Influence of Mold Movement*

It was originally supposed⁸ that these movements would be effective because they would diffuse the direction of gravitational flow within the dendritic array. From the preceding, however, it is now thought that the influence of movement lies in the translation of the bulk liquid across the dendritic front and boundary layer, and it is important to consider the pattern of this translation.

In the experiments described here, the mold was moved in two ways: (i) by rotation about a stationary inclined axis ($\text{NH}_4\text{Cl-H}_2\text{O}$) and (ii) by rotation of the inclined axis about a pivot so that the mold then rotated about the axis in the opposite sense (Pb-Sn). In either case, the result is to cause the upper surface to ride up the sides of the containing vessel, that is, equivalent to a continuous series of rocking movements about axes in the plane of the base, which themselves rotate. Actually, at slower rates, with a liquid of low viscosity, it is the vessel which moves and the bulk of the contents is stationary. Both methods of movement produce an equivalent result inasmuch as the gravitational vector moves continuously about the surface of a cone and the movement of the bulk liquid relative to the planar boundary layer is the same in each case, a rotary scrubbing movement which is the same at all parts of the front, except close to the cylinder walls. The amplitude of translation is determined by how far up the sides of the vessel the liquid moves in each 180 deg of rotation about the vertical axis. If the container were in the form of two parallel sides (Figure 12) of semi-infinite length, the movement could be described entirely by rotation about a horizontal axis through an angle 2θ . The distance moved would then be (Figure 12) $h = D \tan \theta$,

where D is the width of the vessel. Now, within the constraints of a vessel which is not infinitely wide in the other horizontal direction, in particular, a cylinder, there is a point, P , on the side, where there is no translation of liquid, *i.e.*, the liquid level stays the same. Therefore, in the cylinder it is necessary to rotate simultaneously, and the circular rotational velocity is given by $\pi h \cdot \omega$, where ω is the angular velocity (*i.e.*, rpm), or $U = \pi D \omega \tan \theta$.

The preferred argument then, is that such relative movement is sufficient to damp out any perturbation which develops from the boundary layer and/or shear off any plume which has become established, thereby preventing continued channel entrainment and flow. Following the analysis of Figure 12 for the two series of experiments with $\text{NH}_4\text{Cl-H}_2\text{O}$ and Pb-Sn , gives flow rates of $\sim 10 \text{ mm s}^{-1}$ and $\sim 1 \text{ mm s}^{-1}$, respectively, which are most effective in preventing channel formation. While the effect of a shearing movement upon such perturbations has been subject to a linear analysis,¹⁸ it is not clear how far the situation ahead of a permeable front is strictly comparable. It is considered that the order of magnitude difference between the necessary horizontal flow rates in the two cases described here is primarily a reflection of the very different dendritic mesh permeabilities, the very low solid fraction in ammonium-chloride-water making it much easier to supply a developing plume and requiring a correspondingly severe shearing motion to damp out the vertical flow.

Finally, the analysis of the relative flow rate (Figure 12) indicates that it should be proportional to the mold diameter. This dependence was qualitatively confirmed by repeating the tilted rotation experiments with $\text{NH}_4\text{Cl-H}_2\text{O}$ castings in a narrower cylinder of 37 mm bore (half the original). It was found that channels were still present with a rate of 5 rpm about a 30 deg inclined axis, a combination which completely eliminated them in the wider mold.

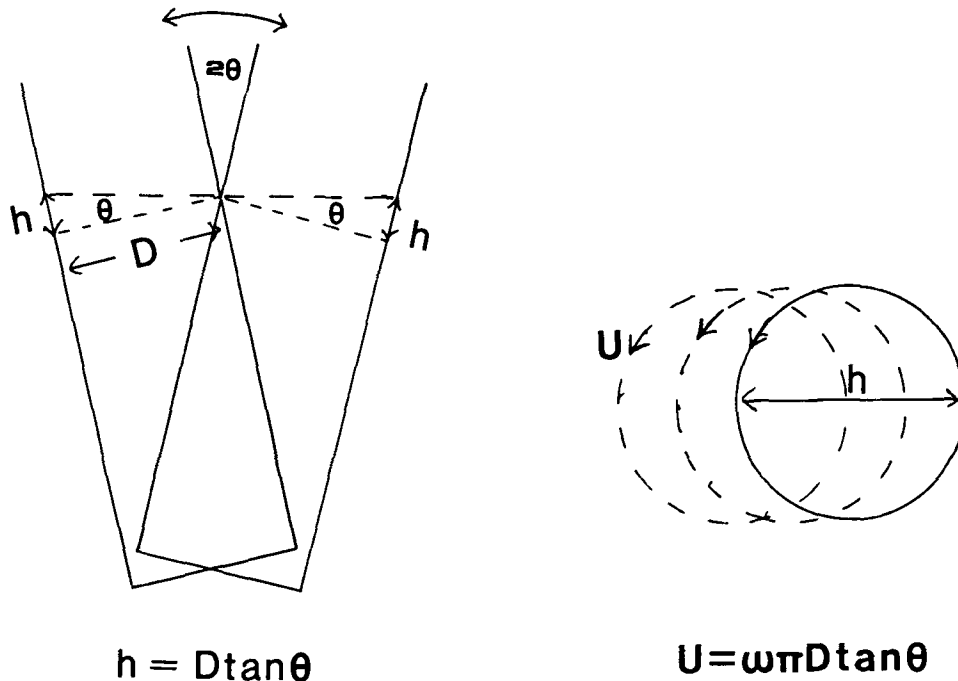


Fig. 12—Showing the liquid displacement, h , which would be caused by rocking a mold of width D about an arc of 2θ . This would be accomplished by 180 deg of rotation about an axis inclined at θ to the vertical and so produce a circular movement relative to any position at the front.

V. CONCLUSIONS

1. In a base chilled configuration, channel flow from the dendritic 'mushy' zone is accompanied by a continuous plume rising through the supernatant bulk liquid. Such flow is maintained by near steady state entrainment of liquid into the growth front between channels.
2. Channels originate, not within the dendritic array at any depth, but immediately ahead of the growth front as a result of perturbation from the less dense boundary layer into the bulk liquid. Such perturbations can be supplied by entrainment to stabilize the developing plumes in a manner which is not possible at a planar solid-liquid interface.
3. The effect of mold movement is to translate laterally the bulk liquid relative to the growth front so that perturbations or pre-existing solute plumes are damped out or sheared off. The particular rotational or precessional movements employed in this work cause the growth front to move in rotary fashion with respect to the bulk liquid, with a velocity which is everywhere the same for given conditions and dimensions.

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