Dendrite Fragmentation and the Effects of Fluid Flow in Castings

A. Hellawell, S. Liu, and S.Z. Lu

In the absence of grain-refining additions to a melt or of any significant heterogeneous nuclei, equiaxed grains can only originate in a casting from primary dendrite fragments. Thus, it is pertinent to understand how dendrites become fragmented and to explain why liquid stirring should appear to "break up" and refine the grain structure. Dendrite fragmentation occurs by local remelting, and fluid flow is important as a dispersal mechanism.

INTRODUCTION

Equiaxed grains in castings can originate from either heterogeneous nucleation (external source) or dendrite fragments (intrinsic source). In the absence of effective nucleation substrates, dendrite fragments are the only source of the equiaxed grain formation.

Five time- and temperature-dependent steps may be identified in a partly solidified casting, starting with the separation of dendritic side arms in the mushy columnar region and ending with the blocking of the columnar front by a network of equiaxed grains (Figure 1).¹ The five steps in the cycle are Step I: columnar growth/dendrite fragmentation; Step II: fragment transport from mushy region; Step III: melting vs. survival; Step IV: growth and sedimentation; and Step V: columnar/equiaxed transition.

An understanding of the physics involved in each of these steps could provide the basis for a modeling sequence to describe the evolution of the grain structure of a casting. Such a program could be written for a time- and temperaturedependent sequence by a series of simultaneous equations, but it is important to get the physics correct and, thereby, make the program predictive. Current modeling procedures (e.g., Dantzig² or Rappaz³) do not attempt to physically identify the sources of nuclei for equiaxed grains, whether intrinsic or extrinsic.

A CYCLE FOR GRAIN EVOLUTION

Step i

The situation depicted in Figure 1 is a breakdown of the original suggestion made by Rosenhain some 70 years ago,⁴ namely, that given some source(s) of primary fragments and a transport mechanism, equiaxed grains can originate without heterogeneous nucleation on accidental or deliberately added substrates. The initial dendritic mushy region is assumed to be the most potent source of such fragments. After the dendrite side arms separate (Step I), the subsequent steps follow.

Step II

Fragment transport to the open liquid region requires fluid flow by natural convection and/or induced stirring. The fluid flow may or may not play some part in the fragmentation process.

Natural convection within and without the mushy region occurs because there are density variations caused by temperature and composition gradients. Such convection is termed thermosolutal. In most alloy castings, the compositiondensity dependence exceeds that of temperature, so that the solute-enriched, interdendritic liquid becomes more or less dense than the open liquid, therefore, tending to sink or rise within the mushy region. Thus, the presence of carbon, silicon, sulfur, or phosphorus in steels causes an upward flow (despite the fact that the interdendritic liquid is cooler), while in alloys such as Al-Cu(Fe,Mn) the reverse occurs, and the more dense, solute-rich liquid tends to collect at the base of a casting or in the sump of a continuous casting.



Figure 1. The grain-evolutionary cycle and transition in castings from intrinsic source.

In the first instance, the upward fluid flow erupts into the open liquid as channel plumes ("A" segregation in billets or freckles in directionally solidified [D.S.] ingots), and these spew many dendrite fragments into the open melt.5-7 In the second instance, the same broadcast transport of fragments is lacking, and this is why it is then necessary to resort to the use of grain refiners to promote the nucleation of equiaxed grains. With liquid stirring induced electromagnetically, the effect may not significantly penetrate into the dendritic array of the mushy zone, but fragments that escape by thermosolutal convection are rapidly swept away into the open liquid. Also, in the mushy region there are millions of potential side-arm fragments, and only a fraction of these need to escape to be effective. In addition, channel plumes may have velocities of up to 0.1 ms⁻¹ in metals,7 so the transport out of the mushy region can be very rapid, even without imposed stirring.

Step III

Fragment melting versus survival depends on the ambient liquid temperature that falls continuously ahead of the columnar front. Since there is a small solute accumulation at the dendritic front, the freezing point is depressed by a few degrees (e.g., <5 K). Therefore, it is possible for the entire open liquid to become slightly supercooled. Dendritic fragments typically have sizes around 10-50 µm and are moving with respect to the liquid. At temperatures above the liquidus, these particles dissolve increasingly rapidly by liquid-solute diffusion up to a temperature where they melt without composition change.8

Estimates of probable lifetimes for the dissolution of such particles are less than 10 s and when they reach their melting temperatures, lifetimes cannot be expected to exceed ≈ 1 s. Therefore, such particles are unlikely to survive long enough to become sources of equiaxed grains until the liquid becomes supercooled, or unless they are swept into cooler regions as by imposed stirring. Thus, in a rapidly stirred rheocasting with fluid-flow rates as high as 1 ms⁻¹, a particle could be circulated around the liquid pool in less than 0.1 s and have a much higher chance of survival than it would in a quiescent liquid. This may be



Figure 2. An illustration of the assumption that a free dendrite during growth/sedimentation is surrounded by a quiescent liquid envelope with a little higher concentration than the bulk.

one reason why liquid stirring favors a refined equiaxed structure.¹⁰

Of the five identified steps in the cycle, this question of particle survival is the most difficult to quantify. One possible approach to the problem is in terms of particle population density based upon a "birth rate" (particle supply from the mushy region) versus a temperaturedependent "death rate" (from dissolution and melting) expressed rather like a demographic problem.¹ But this is an arbitrary formality and presently lacks a satisfactory physical analysis.

Step IV

If and/or when a surviving particle reaches liquid at a temperature below the liquidus, it begins to grow dendritically and (with solid density exceeding that of the liquid by a few percent) it tends to sink, subject to convection currents or induced stirring. For a quiescent liquid, this is a situation that can be analyzed with some confidence11 by combining the temperature- and concentration-dependent dendritic growth rate with a sedimentation rate based on a modified version of Stokes' equation. Such crystals grow as they fall and, in principle, should accelerate. But, although the solid volume is increasing, the actual solid fraction within the envelope or outline of the crystal is small (e.g., <5 percent), and it is not known how liquid flows through or around such a filamentary crystal (Figure 2).

One approach used by Jang and Hellawell¹¹ assumes that the interdendritic liquid atmosphere is essentially quiescent; it also has a slightly different density than that of the bulk, because it is slightly enriched in solute. The shape is not quite spherical because of the orthogonal dendritic outline, but this is a minor factor in Stokes' equation, and, in any case, such crystals often rotate as they move through the surrounding area. An analysis of this form has achieved acceptable agreement with laboratory experiments using the ammonium-chloride water analog for metals and can, by extension, yield grain sizes for continuously cast steel billets that are of the correct, observed order of size. Beckermann¹² has adopted a similar model

There is an extra quirk to this problem in that such dendritic crystals might reproduce themselves by producing further side-arm fragments. This is an extra complication and a moot point. It could be that with very small superheats yielding almost entirely fine, equiaxed crystals (a result that Chalmers called big bang nucleation¹³), a process of multiple fragmentation occurs, rather like a roman-candle firework. It is also possible, however, that the necessary fragments are swept off the fine dendritic chilled crystals that nucleate on the mold walls. The answer to this question is not known as it all happens within a few seconds.

Step V

In the end, there is a cloud of growing, sinking, dendritic crystals, and when the number and size of these exceed a certain level, continued growth of the initial front is blocked. This is the columnar-to-equiaxed transition. Of course, there is a geometrical problem to be addressed, according to the growth direction of the columnar front, whether horizontal or vertically upwards. Either way, the basic analysis concerns how many falling grains of what size must reach the front in order to prevent continued growth around and between the arriving obstacles. This can be addressed with varying degrees of sophistication, but to a first approximation, the necessary criterion is that the arrival of obstacles be sufficient to half cover the front in less time than that front can grow around and/or between them. Analysis, therefore, includes the number of crystals of given size arriving per unit area of front in a given time at a given angle versus the original rate of advance of the columnar front; it is a soluble problem.14

DENDRITE FRAGMENTATION

Everything in the grain evolution cycle was predicated on the existence of dendrite fragments being available within the initial columnar (or chilled) zone—







Figure 4. The organic analog of SCN-5.5H₂O (wt.%) system showing that under steadystate conditions (V and G are constant), dendrites ripen and side arms become necked at the junctions with primary stalks, but they do not detach and fragment unless the growth undergoes a deceleration or temperature rises.

Step I. It must be stated that metallic dendrites do not break mechanically in the course of a casting operation. They may be plastically bent, but some simple estimates¹⁵ based on bending moments and fluid-flow calculations indicate that this will be a rare event.

There are two situations to consider. Either the dendritic crystals are attached to the mold walls as a permeable array or relatively isolated dendritic crystals are floating about in open liquid or being swept along with the liquid in natural or induced flow patterns. In the first instance, the liquid can only move through the array; in the second, most of the liquid probably flows around the envelope of the crystal. These isolated filamentary crystals with liquid atmospheres have densities very similar to the bulk liquid and easily move with any currents, like feathers in air. They may occasionally jostle each other as they move along, but the chances of a sufficient impact to plastically deform them is remote. There is more concern with interdendritic flow within the columnar array, and if the estimates are correct, this is also rarely sufficient to cause more than elastic flexing of side arms. Other mechanism(s) must operate, and it is relevant to consider dendritic array/ growth and ripening behind the growth front.

In Figure 3, three dimensions are important: the primary dendrite spacing, λ_1 ; the radius of curvature of the dendrite tips, r; and the initial secondary arm spacing, λ_2^0 . The last of these is generally a multiple of the oscillating tip radius, which is only marginally stable.16 As the front advances, secondary arms coalesce and shrink or grow, so that with ripening, secondary arm spacing increases continuously as λ_2 . Under steadystate conditions of growth rate (V) and temperature gradient (G), the product r²V is approximately a constant, and a small range of stable primary λ_1 is also preferred.¹⁷⁻²⁰ Side arms do not detach from the primaries under these conditions, but the roots or nodes become

necked (Figure 4) because of excess solute rejection from both primary and secondary arms. These necks do not separate unless the growth conditions (V and G) change, which they do. To cause detachment, either the local temperature and/or the local solute concentration must rise so that the arms melt off at the necked roots. Whether convective flow within the array contributes to the melting process is another arguable point, but it seems more probable that such flow is primarily important as a transport mechanism for particles that detach for other reasons.

In a casting, the growth rate of a columnar front decreases continuously; if the mold metal interface temperature were constant, it would do so parabolically as the temperature gradient decreased. Taken separately, a decrease in the growth rate causes the radius of curvature of dendrite tips to rise (a rapid response) and the preferred primary spacing λ_1 to increase. But the latter adjustment is slow and requires that dendrites periodically fall back and stop growing (Figure 5) so that at any instant the primary spacing is always smaller than it would be under steady-state conditions at any given velocity.21-22

There is a similar reverse hysteresis in the primary spacing adjustment with acceleration.^{23–24} For this reason, the liquid solute concentration within the array is continuously rising above what would have applied at an earlier instant. Here is a first contribution to accelerated local melting. Second, still supposing that the mold interface temperature re-



Figure 6. The percentage of secondary arm detachment versus time with various deceleration (a) in the analog casting of SCN-5.5H₂O (wt.%) system.

mains constant, a falling temperature gradient at the growth front also favors an increase in primary spacing and tip curvature. This combination of transient conditions acts to promote local melting at the secondary-arm roots. Experiments with transparent analogs (SCN-H,O) show that the fraction of side arms that become detached is proportional to the deceleration rate and can be as high as 80 percent (Figure 6), which means that the number of potentially available side-arm fragments is enormous. (e.g., consider 1 mm³ of mushy array, where $\lambda_1 \cong 50 \,\mu\text{m}$ and $\lambda_2 \cong 10 \,\mu\text{m}$; the number of side arms in this volume is then $400 \times 20^2 = 1.6 \times$ 10⁵ mm⁻³).

In a real casting, the metal/mold interface temperature is not constant. In most individual castings (e.g., billets), the mold temperature rises, especially in a sand casting, so that the temperature within the mushy region close to the mold behind the columnar growth front

> will actually be rising, and some side-arm melting/detachment is inevitable, even without the previously identified solute enrichment.

In a continuous casting, the situation is the reverse because the ingot surface temperature falls as the metal moves through the mold and briefly rises as the strand enters the water-spray region before falling again. In such cases, the fraction of detached side arms will be smaller and depend more critically upon the rate of deceleration of the columnar growth front. There will be a trade-off here between the falling temperature at any position in the mushy region and the solute enrichment associated with decelerating growth.

The essential point is that fragmentation or crystal multiplication²⁵ takes place by local remelting, and fluid flow does not break-up the structure in a mechanical sense; perhaps "disperse" would be a more accurate description of stirring action.

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ABOUT THE AUTHORS

A. Hellawell earned his D.Phil. in physical science at Oxford University in 1956. He is currently emeritus research professor at Michigan Technological University. He is also a member of TMS.

S. Liu earned his M.S. in materials science and engineering at Northwestern Polytechnical University, China, in 1989. He is currently a Ph.D. candidate at Michigan Technological University.

S.Z. Lu earned his Ph.D. in metallurgical engineering at Michigan Technological University in 1986. He is currently a research associate professor at Michigan Technological University. He is also a member of TMS.

For more information, contact A. Hellawell, Department of Metallurgical Materials Engineering, Michigan Technological Institute, 1400 Townsend Drive, Houghton, Michigan 49931-1295; (906) 487-2261; fax (906) 487-2934.



Figure 5. The organic analog of dendritic growth with deceleration (from 30 µms-1 to 3.3 µms-1) in the SCN-5.5H₂O (wt.%) system showing that the primary spacing adjustment exhibits a hysteresis in response to the velocity decrease. Note that in this picture, the growth velocity is already approaching the lower end (V = 3.3 µms⁻¹).