A Short History of MIT Studies on Fluid Flow in Solidification, 1952- 2009

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The Short History

My first published paper [1] was in 1956, as a newly minted assistant Professor at MIT. The paper comprised a simple experiment on effect of fluid flow on solidification. The abstract of the paper began as:

"The grain structure of a casting is commonly described in terms of alloy composition, cooling rate, and nucleation. It is shown here that a fourth variable must sometimes be considered, the flow of liquid across the solidifying interface…."

The paper was an outgrowth of graduate thesis work conducted several years earlier with one of my two college roommates, fellow student Edward Hucke. It showed that metal flowing past columnar grains causes the grains to grow in the "upstream" direction, and explained the result in terms of the then quite new work of Bruce Chalmers and his students on "supercooling" in dendritic growth. Figure 1 shows two castings we produced. Both castings had a rectangular cross section and were bottom chilled, filled from one end. In the case of the casting at the top, the end opposite the gate entrance was left open so the metal flowed out the far end during metal pouring. We were delighted to see the columnar grains that grew in the first case pointed nicely upstream. The casting at the bottom of the figure had its end plugged so filling occurred as in a usual casting; the result was a largely equiaxed grain structure. Of course, if we had looked at the structure of the solidified metal that flowed out of the open end of the mold we would have found a very fine grain indeed, but that was a finding that would have to wait decades, until the advent of semi-solid processing and flow down a sloping chill face.

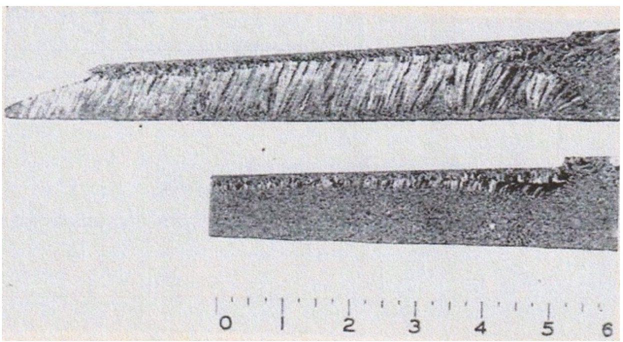


Figure 1. Bottom chilled commercial purity (99.85%) aluminum castings, etched longitudinal section. In the upper casting, metal spilled out the left end during pour. In the lower casting, the end was plugged.

My interests in the following 5 years were largely directed to improving the mechanical properties of cast metals, especially aluminum alloys. We learned in this period of the importance of control of dendrite arm spacing, interdendritic precipitates and interdendritic porosity. We recognized that to achieve soundness, liquid metal must be caused to flow interdendritically to feed solidification shrinkage, and so interdendritic fluid flow, Figure 2, became an important part of our research. Interdendritic porosity is reduced by both minimizing dissolved gas content and by minimizing the pressure drop as liquid flows through the dendrite interstices to feed shrinkage. This pressure drop is reduced by achieving short distances between risers and chills, as in Figure 3.

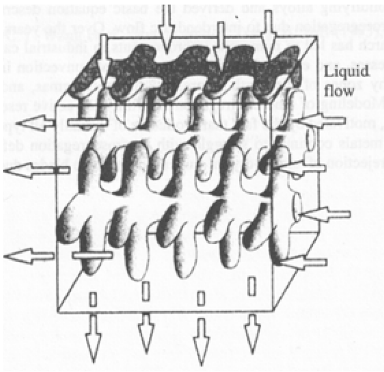


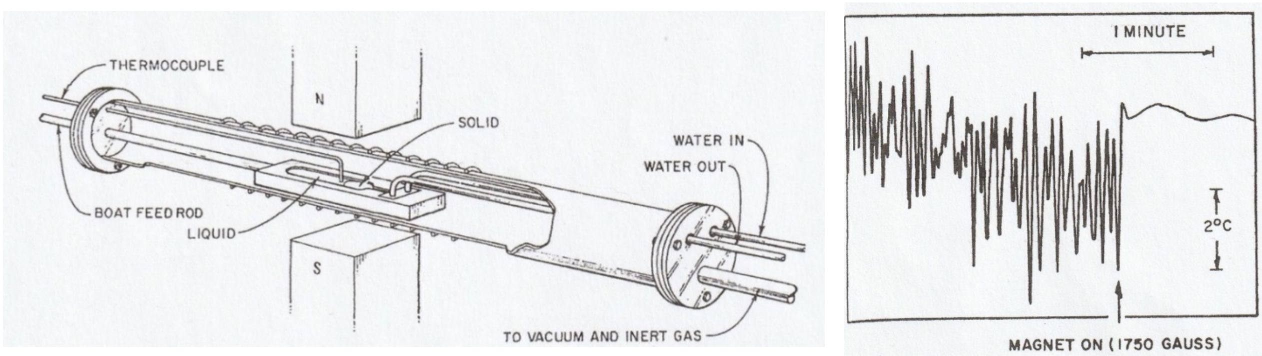
Figure 2. Schematic of interdendritic fluid flow through a fixed volume element in the liquid solid region.

Figure 3. Left) Drag half of a sand mold for a "premium quality" aluminum sand casting, showing chills embedded in the mold. Right) finished casting.

Techniques [2] for accomplishing the foregoing in actual castings were introduced industrially as early as 1960, and became the basis for Federal Specification MILA21180. The current commercial embodiment of this specification, now widely employed, is AMSA21180C.

As is now widely recognized, this interdendritic flow is responsible for macroscopic composition differences across a casting or ingot; i.e., macrosegregation [3]. In simple castings, the interdendritic flow is usually primarily in a direction opposite to that of isotherm movement. In large ingots, or in unidirectionally solidified castings (e.g., columnar turbine blades), flow can sometimes be locally in the same direction as that of isotherm movement, and if sufficient this flow results in the "freckles" in the directionally solidified casting and the streaked segregates (e.g., "A segregates") in ingots.

The early 1960's began a period of rapid growth in the computer industry, and with it much research interest in semiconductor crystal growth to achieve crystal perfection and precise and uniform composition. A problem that plagued researchers was periodic compositional variations, termed "solute banding." There were differences of opinion in the field as to whether the banding was caused by an interface kinetic effect, or by mass transport in the diffusion boundary layer at the growing solid interface. To test this, graduate student Harvey Utech built apparatus to solidify In-Sb alloy in a constant D.C. magnetic field, Figure 4a. The expectation was that the magnetic field would dampen convection and would thereby reduce any banding caused by variations in mass transport across the boundary layer. The effect on convection was pronounced, as shown in Figure 4b. Before application of the magnetic field, a sensitive thermoocuple in the melt picked up the thermal fluctuations caused by thermally driven convection. After application of the magnetic field the fluctuations disappeared. The combined fluctuations of fluid flow and temperature were anticipated to be a direct cause of banding, and examination of the solidified ingots showed that the solute banding disappeared when the temperature fluctuation was eliminated [4,5]. Others soon showed that a steady D.C. magnetic field, by reducing convection at the tips of growing columnar dendrites, reduced the tendency for dendrite arms to "break off, forming an equiaxed structure.



(a) (b)

Figure 4 (a) Utech thesis apparatus for study of effect of DC magnetic field on convection in crystal growth. (b) Thermal fluctuations are shown in the liquid ahead of the flowing solid interface. Upon application of a 1750 Gauss DC magnetic field, the fluctuations are greatly reduced.[4,5]

The foregoing studies all pertained to the effects of relatively mild convection on structure, and in the late 1960's we undertook to study effect of very high convection on structure. This evolved into David Spencer's doctoral thesis [6] in which he solidified tin-lead alloy in the annular space between two counterrotating cylinders, Fig. 5, with the resulting invention of what we now term "semi-solid processing".

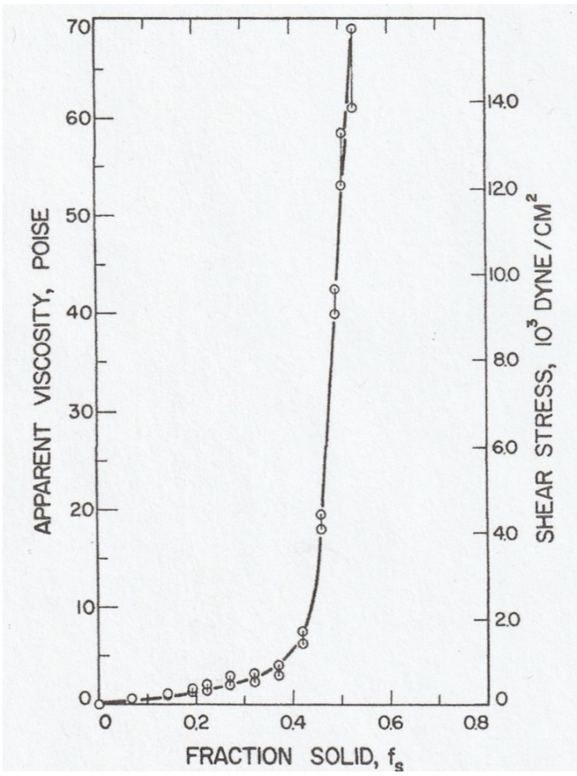


Figure 5. Measurement in David Spencer's thesis of viscosity versus fraction solid in a Sn-Pb alloy.[6]

My last papers relating to semi-solid processing, published in 2009 [7,8], were with my second (and long a former) college roommate, David Ragone. The papers were a quantitative thermodynamic and solidification study of the "puddling" process, a semi-solid processing method that provided inexpensive wrought iron at high volumes through the 19th century. It was a mainstay of the industrial revolution until it was gradually replaced by modern steelmaking methods at the beginning in the 20th century.

Figure 6 is puddling from a metallurgical and solidification perspective. The starting point in practice was a cast iron of roughly 3% carbon, with silicon and other elements present. However, in Figure 6, the process is modeled more simply by assuming a binary starting alloy of only iron and carbon. The alloy is first melted, further heated, and then cooled slightly with some carbon removal by evolution of carbon monoxide, following the line OAB in Figure 6. At point B solidification begins and the evolution, now rapid, of carbon monoxide heats the metal exothermically while the average carbon composition of the solid-liquid mixture carbon content decreases along the line BC′. During this period, the iron worker "poles' (stirs) the metal vigorously. The puddling continues until it can no longer be "worked" by the puddler. The metal is then largely solid at the desired low carbon content, C′. The remaining "ball" of metal is then removed from the furnace and pressed to remove as much slag as possible. The main steps of puddling are pictured schematically in Figure 7 (except the figure does not make it plain that during the step BC′ the metal is semi-solid).

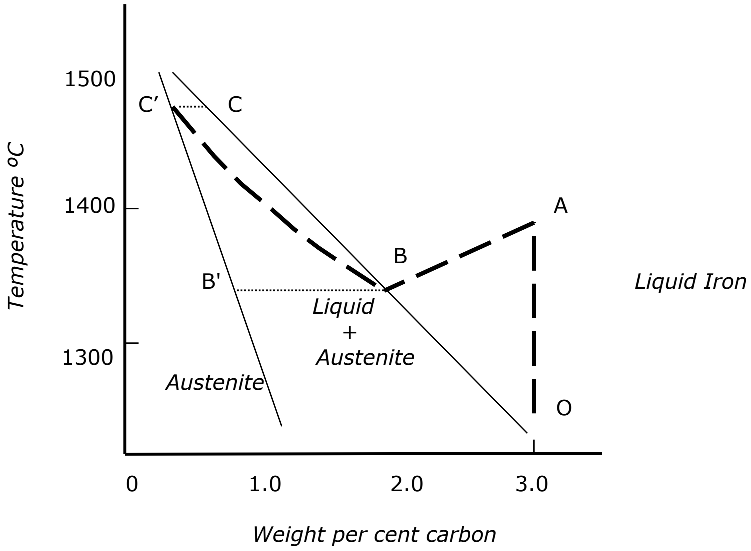


Figure 6. Puddling from a metallurgical and solidification perspective[8]

Figure 7. Schematic Illustration of Puddling

Concluding Remark

Solidification science and engineering are critical to processing of a wide range of materials for a wide range of applications. I have been happy to work in one aspect of that broad field for seven decades, mostly on metals in the semi-solid state. I wish you all success and satisfaction in your own careers in this field and beyond, and thank you for this opportunity to reminisce a little.

References

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