DENDRITE MORPHOLOGY

by

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ABSTRACT

A study has been made of the dendrite morphology of an aluminum-copper alloy solidified under widely varying conditions. Initial work was on the structure at chill surfaces. If suitable casting techniques are employed, this structure is revealed by shrinkage during the later stages of solidification. The primary dendrite traces (or stalks) visible on the chill surface are either the intersection of a {100} dendrite plane with the chill plane and/or the projection of a <100> direction onto the chill plane. The secondary "branches" are usually at 90° to the primary, independent of dendrite orientation. This is due to the growth of a {100} dendrite plane as the primary dendrite element, in conjunction with the projection of a <100> direction of identical indices as secondary dendrite element, or vice versa. When the secondary branches are not at 90°, some other combination of {100} plane or projection of <100> direction occurs with an unlike indexed plane or projection of direction (and can only be true of a grain off the <100> zones). The growth of {100}'s dendritic planes and projections of <100> directions has been examined as a function of orientation of the grains.

In spite of preferred growth of the {100} planes and <100> directions, very little preferred orientation or texture exists in grains at the chill. The evidence indicates that grains of all orientations have an equal probability of being nucleated under the conditions of the experiments performed.

A fine-grained structure is produced without grain refining additions in chill castings, at the chill face, if flow of metal occurs during solidification. Several mechanisms of grain refinement by convection are discussed to explain this effect.

Bent dendrites are observed on the chill surface. X-ray analysis shows that the crystal lattice of bent dendrites is concave toward the chill surface. An explanation which correlates these two phenomena is suggested.
Work was also conducted on the morphology of columnar grains. One major feature of columnar dendrite morphology, namely that a basic form of the dendrite is a sheet-like structure, has appeared before in the literature but has been largely lost in the classical conception of "rod-like" dendrites. Another major feature is the development of a substructure within the grains consisting of largely independent "dendrite elements".

The development of dendrite elements is examined in some detail as a function of cooling rate, and a theory proposed to explain the development of the substructure in terms of the sheet-like morphology. The formation of the dendrite element substructure results in anisotropic heat conductivity in the solid and the solid plus liquid zones. This effect has been used to explain the development of a preferred orientation during columnar growth. The influence of dendrite orientation on columnar morphology has been studied using seeded crystals. Orientations studied were \langle100\rangle, \langle110\rangle and \langle11\rangle. The results correlate with the columnar ingot studies, showing development of \{100\} dendrite planes. Also, they indicate the stability of grains having one \{100\} plane parallel to the heat flow direction.

Definite boundaries of high segregation exist within the dendrite; these are boundaries of the dendrite element. Segregation within dendrites has been studied by means of the electron beam microprobe. Studies of air and water chilled columnar dendrites, of air chilled columnar dendrites quenched with water chilling, and of equiaxed dendrites removed from their liquid bath by means of lead displacement have given results consistent with the theoretical work of Brody and Flemings, showing that significant diffusion occurs in the solid during the solidification of aluminum-copper alloy.

Study of the equiaxed dendrites also show that their morphology is essentially the same as columnar dendrite morphology, except that the latter are extended in one \langle100\rangle direction.

The development of "double orientation" in columnar dendrites has been studied. In this case, the dendrites develop a preferred texture in two perpendicular directions as a result of growth from two chills. Thus, all grains have nearly the same orientation. A figure of merit has been derived to measure the extent of double orientation. A successfully doubly oriented casting was 40 per cent doubly oriented: a casting in which no attempt was made to achieve double orientation was 8 per cent doubly oriented.

Thesis Supervisor: Merton C. Flemings

Title: Associate Professor of Metallurgy
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I. GENERAL INTRODUCTION

Almost all commercial metals are solidified at some time during their processing. Under normal conditions, solidification of metals is dendritic; therefore, the vast majority of all metal produced undergoes dendritic freezing. The resulting casting or ingot may subsequently be worked and annealed, but the effects of the solidification process are manifest in products far removed (in their processing history) from the liquid-solid transition. Studies on dendritic solidification comprise, therefore, an important portion of materials processing research, a portion with broad technological implications.

Dendrite Structure

The most obvious feature of a casting or ingot, beyond its external shape, is its dendritic structure. The dendritic structure may often be seen on the surface or in shrinkage cavities of a casting; it may also be examined on a cut, polished and etched section from a casting or ingot. There are, therefore, several ways of gaining information about the shape or morphology of dendritic crystals. Nevertheless there has been some confusion concerning dendrite morphology.

Dendrite Character

First of all, confusion arises from the word "dendrite" itself. "Dendrite" is derived from the Greek "dendrites" meaning "of a tree". The implication, which is often expressed, is that a dendrite is "tree-like". It is true that a dendrite is tree like in some ways, but there are noteworthy differences. There are two major similarities between a tree and a
dendrite. (1) Both are highly involuted structures, (2) both are quite "disconnected" or "branching" structures. Dendritic disconnection is discussed later. The dissimilarities between a tree and a dendrite include: (1) a tree is never crystallographic in its growth directions unlike a dendrite, (2) a tree does not almost totally fill space after its growth is completed, (3) detailed structure of the two is entirely different.

Two and Three Dimensional "Dendrites"

Another area of confusion arises due to the varying methods of observation of dendrites. For example, the traces seen on the surface of castings or etched specimens are often called dendrites; this is a two-dimensional aspect of a three-dimensional structure. The following are some of the definitions implicitly given for the term dendrite:

A. Three-dimensional structure (conceptual and/or decanting)
   1. dendritically solidified grains, totally solid
   2. dendritically solidified grains, partially solid (the skeleton)
   3. a major "branch" of a dendritically solidified grain, totally solid
   4. a major "branch" of a dendritically solidified grain, partially solid

B. Two-dimensional structure
   1. the intersection of a dendritically solidified grain with the plane of polish
   2. the intersection of a main "branch" of a dendritically solidified grain with plane of polish
   3. the differential etching part of an intersection of a dendritically solidified grain with the plane of polish (interpreted as an early stage in growth of the dendritic skeleton)
4. the differential etching parts of an intersection of a main "branch" of a dendritically solidified grain with the plane of polish

For the purpose of this thesis,¹ a dendrite is that portion of a grain that is solid at any stage of solidification. The dendrite may be composed of completely contiguous parts or it may be subdivided into largely independent "dendrite elements", as described later. These elements are of crystallographically related orientation, i.e., in cubic systems the primary axes of the elements are approximately parallel or perpendicular to each other since they have grown from a single nucleus.

Classically, dendrites have been represented as "rods". For this conception, rods that form the primary axes of dendrite elements are termed primary growth forms (primary rods, primary axes, primary dendrite arms, primary projections). Rods which grow perpendicular to the primary rods are secondary growth forms (secondary rods, secondary dendrite arms). Tertiary and quaternary growth forms may also be present (see Figure I-la).

It will be clear that in some systems and at some stages of growth, dendrite morphology is at least partly plate-like or sheet-like. Interstices between primary and secondary "arms" tend to "fill in" at an early stage of solidification (or in some cases it may be that well developed arms never form); the sheets which form in this way are termed primary plates or primary sheets (see Figure I-1b). Sheets oriented parallel to secondary and tertiary arms and perpendicular to both primary sheets are termed secondary sheets; sheets parallel to two tertiary arms are tertiary sheets.
Figure I-1: Schematic picture of a dendrite, showing growth forms and dendrite elements. (a) Two-dimensional representation, showing dendrite elements, (b) part of a dendrite element, showing the relationship between terminology in rod-like and sheet-like dendrites.
Dendrite Growth Directions

Some confusion is caused by the relationship between dendrite growth form growth direction, grain (boundary) growth direction, and preferred orientation. A single dendrite tends to grow with projections (axes) in low index crystallographic directions. These are \( <100> \) directions for FCC materials, for example. But there are exceptions to this rule under special conditions. For example, Miksch\(^2\) has shown that in ice, dendrite arms may be forced by convection to grow 20° from the usual low index direction. Also, metal dendrite growth forms growing toward one another have been observed to deflect from each other, due to solute rejection from each affecting the other.\(^3\) Finally, it is obvious that in order to fill space elements must grow away from low index directions near the end of their growth.

In the three examples above, the dendrite axis growth direction deviates from a low index crystallographic direction, while the crystal lattice of the grain remains macroscopically undistorted (i.e., without major change in orientation over large translations through the lattice). Lindenmeyer\(^4\) has investigated the growth of ice dendrites under varying conditions. When grown on a substrate, the dendrite axes are bent. The bend corresponds to a change in orientation of the crystal lattice and occurs in such a way as to align the basal plane to the substrate.

Studies of the growth of a group of dendrites (or dendrite elements from different grains) into a columnar structure\(^5\) show that the grains often tend to have a preferred orientation. The direction of growth of the dendrite elements tends to become aligned with the heat flow direction. For example, FCC grains tend to be aligned with their \( <100> \) directions within 10 - 20° from the heat flow direction. It should be emphasized that under normal
conditions the individual dendrites grow with their axes exactly in a (100) direction, which is therefore within 10° - 20° from the heat flow direction. As a consequence of the development of a preferred growth direction, the grain boundaries also tend to be aligned perpendicular to the heat flow direction. However, Flemings et al.⁶ have shown that a flowing stream causes the grain boundaries to be tilted upstream. This is due to a washing away of solute from the upstream side of the dendrite elements. The dendrite growth forms, however, continue to grow with a preferred orientation perpendicular to the mold wall. Palmer⁷ has shown that the preferred orientation itself will point upstream if the fluid is flowing before the columnar grains have started to grow. In this case, the grain boundaries are aligned in the direction of preferred orientation, which is the dendrite growth form growth direction, but which may not be in the heat flow direction.

Dendrite Structure Study - Historical

Since "dendrites" can sometimes be seen on the surface and in risers of castings, one would expect that dendritic structure must have been observed almost as soon as metals were cast. The first description of the dendritic skeleton, however, was not recorded until 1775, by a French ironmaster, Grignon. He felt that dendrites are built up by the accretion of regular rhombs upon each other in successively decreasing size. This and other imaginative ideas and his accompanying drawings were an idealized abstraction from observation.⁸ (See Figure I-2.)

The first systematic investigation of dendritic growth seems to have been made by Professor D. K. Tschernoff. In his paper of 1880 "On the Structure of Cast Steel Ingots"⁹ he provided both pictures of dendrites
Figure I-2: Grignon's sketch of a dendrite (1775).
observed in cast steel (see Figure I-3) and his often copied revision of Grignon's abstraction (Figure I-4). A cursory examination of these drawings and photographs shows a wide divergence between the abstraction and the observation. Tschernoff remarked: "When examining individual crystals under the microscope, we observe that every crystal is a skeleton of a drawn out square octahedron. Besides the shoots in the direction of the octahedral axes, or of the first order, there are secondary and tertiary shoots, frequently forming regular reticular beams of an octahedral crystal."

At about the same time (1878) Martens published a drawing of a cast iron dendrite, very similar to Tschernoff's observations (see Figure I-5) but with a more accurate written description. They are, Martens says in substance, skeletons of octahedra. They are composed of parallel teeth, attached normally to the principal axes in two planes at right angles to each other. This formation continues frequently to the third and fourth degree.¹⁰

Osmond typified nineteenth century thought on dendritic structures, strongly influenced by the idea of an ideal idiomorphic octahedral crystal toward which dendrites tend to grow. "In short, all steel crystals, whatever their dimensions and their perfections, are made up like those of cast iron, by successive ramifications parallel to three rectangular axes; their limits are heaps of interlocked octahedra, whose principal axis coincides with the three axes of the system. The extremities are sharper than those of a true octahedron, but as these extremities represent the envelope of a row of interlocked crystals, there is no reason why each individual crystal should not be a perfect octahedron, although it is difficult to ascertain it by direction measurements."¹¹
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Figure I-3: Tschernoff's 15-inch steel dendrite.
Figure I-4: Tschernoff's sketch of a dendrite (1880).

Figure I-5: Marten's drawing of a cast iron dendrite (1878).
Osmond had earlier pioneered the etching of polished sections. By the turn of the century, metallography had joined shrinkage and fracture studies in the study of dendritic structures. Although many excellent photographs were taken earlier, it was not until 1929 that a new interpretation was given to the metallographic evidence of dendritic structure.

In his study of crystallization of 1.10 per cent carbon steel ingots, Hultgren concluded that the main branches (of columnar grains) represent "sections through planes or plates". Later he continues: "Turning to the structure of the individual elongated crystals, it has been shown that a great number of parallel cruciform stems or plate crosses may develop within each crystal at an early stage in its life. In transverse sections such plates appear as lines forming crosses, in longitudinal sections as so-called beams, and in oblique sections as bands . . . It would appear that the individual plates or stems belonging to each crystal during their growth may often lose connection with each other - that is, become separated by the melt over a considerable length". Hultgren used the idea of an idiomorphic octahedron as one of the bases of his explanation of the morphology of dendrites. The influence of concentration gradients on the assumed octahedron resulted in "... a star of six branches radiating from the nucleus, each branch having a cruciform cross-section and tapering towards the points". He did not explain, however, how concentration gradients could influence an octahedron that had never existed.

Thus by 1929 some important features of dendrite structure had been investigated and published. Grignon's conception of a rod-like dendritic
skeleton had been questioned by Hultgren. Still unresolved was the relationship of the equilibrium idiomorphic crystal to the dendritic crystal. Recent work on dendrite structure is discussed in the chapters which follow.

Dendrite Segregation

The alloy selected for this work was ungrainrefined aluminum alloy 195 (high purity aluminum-4.5 per cent copper). The phase diagram of aluminum-copper is given in Figure I-6; the composition of 4.5 per cent copper alloy is shown on the diagram. It has been shown\textsuperscript{14,15} that this alloy solidifies over a wide range of freezing rates in reasonable agreement with the predictions of the classical equation for normal non-equilibrium solidification (perfect diffusion in the liquid, no diffusion in the solid, constant partition coefficient). The predicted segregation curve calculated from this equation $C_S = kC_0(1-f_S)^{k-1}$, is plotted in Figure I-7, where $C_S$ is the composition of the solid, $C_0$ is the initial liquid composition, $f_S$ is the fraction solid, and $k$ is the equilibrium partition ratio. The composition of the first solid according to the assumptions should be about .7 per cent copper and this minimum should be found somewhere in the fully solidified dendrite. Michael and Bever,\textsuperscript{14} however, in their study of segregation by the autoradiographic technique, found a minimum of 2 per cent copper in 4.06 per cent copper casting. They suggested that diffusion in the solid might be responsible for the discrepancy. Brody has made detailed calculations which support this view.\textsuperscript{16}
Figure I-6: Phase diagram of the aluminum-copper system, showing the alloy used in this thesis, 4.5 per cent copper (A.S.M. Metals Handbook, 1948 edition).
Figure I-7: Segregation curve for aluminum-4.5 per cent copper assuming perfect diffusion in the liquid, no diffusion in the solid, and equilibrium at the solid-liquid interface. Applied to microsegregation across a dendrite growth form.
Selection of Alloy for Study

Aluminum-4.5 per cent copper was chosen for study in this work for several reasons. (1) Because of its moderate melting point, it is relatively easy to handle. (2) Aluminum-4.5 per cent copper produces a structure which is readily interpretable. It is easily polished (compared to low melting point metals), and readily shows coring and eutectic structures. With the etchant used, the first solidified (low copper) solid etches lightly, while the copper rich solid etches darker. (3) The brittle eutectic, solidification shrinkage, and gas solubility of this alloy result in segregation, porosity, and structural properties typical of other metals. Its FCC crystal structure and resulting dendrite morphology is similar to many other metals. For these reasons, it may be useful as simulator of solidification in more refractory metals. (4) 195 alloy is a commercial alloy. Much experience has been gained in its use, and data is available on its casting characteristics.
II. CHILL GRAIN STRUCTURE AND PREFERRED ORIENTATION

A. Introduction

1. General.

Ingot and casting solidification often is described in terms of the formation of three zones: \(^{17-19}\) the chill zone, the columnar zone and the equiaxed zone; sometimes all of these "zones" may be present, sometimes only one or two of them. Only the chill zone is discussed in this chapter.

The classical explanation of chill zone formation is as follows: \(^{17}\) Liquid metal enters a cold mold. The liquid in contact with the cold surface rapidly cools to its nucleation temperature and crystals form. Nucleation may occur on the mold wall or in the liquid, depending on the nucleation temperatures for catalysts contained on the mold and in the liquid respectively. For nucleation in either location, grain size is inversely proportional to the number of catalysts. The thickness of the chill zone depends on the depth of the supercooled region when nucleation occurs. Walker states that "... In most instances nuclei form on the mold surface only, and the chill zone is limited to the surface grains in the casting." \(^{17}\)

2. Preferred Orientation.

There are several possible definitions of "preferred orientation". The basis can be the number of grains or the area of grains in a cross-section. If all grains are within \(10^\circ\) of a certain orientation, for example, a preferred orientation is present independent of the basis of measurement. In this case, grains of other orientations are totally absent, so this could be called an "absolute preferred orientation". If all orientations are
present but their density is low except near a certain orientation, then a
preferred orientation is present on a grain number basis. This might be
called "cluster preferred orientation". Most deformation textures have
preferred orientations of this type. A third possibility for preferred
orientation exists even when grain orientation is random. Grains of a
certain orientation may be larger than the other grains. This would give
an "area base preferred orientation".

Edmunds\textsuperscript{20a} has studied the development of preferred orientation in zinc,
cadmium and magnesium. In zinc and cadmium he found that the surface region
had a (0001) texture (parallel to the chill surface), while a (10\bar{1}0) texture
developed in the columnar region. In his discussion of the article,
Greninger\textsuperscript{21} suggested that "... dendritic growth and preferred orientation
in castings must be intimately allied..." In their study of dendritic
growth, Weinberg and Chalmers\textsuperscript{23} observed that the dendritic growth directions
in zinc are the six \{10\bar{1}0\} directions in the basal plane and less well
developed axes in the [0001] direction. Using Greninger's suggestion and
Weinberg and Chalmers' results, Walton and Chalmers\textsuperscript{23} explained Edmunds'
surface and columnar textures. They reasoned that the fast growth \{10\bar{1}0\}
directions are in the basal plane, so that a "... small crystal in a super-
cooled melt will grow as a flat plate parallel to the basal plane. The
nuclei that grow parallel to the mold wall would produce larger grains than
nuclei with other orientations. These large grains would have a basal plane
parallel to the mold wall and a preferred orientation could be observed to
exist on the ingot surface." This would explain the observed chill texture.
"If solidification continued with a dendritic interface, however, the pre-
ference would now be for grains with a \{210\}...\{10\bar{1}0\}..."
direction in the growth direction." In this way, Walton and Chalmers explain the texture change from the chill region to the columnar region.

Similar reasoning might also apply to cubic materials. In this case the $\langle 100 \rangle$ directions are the dendrite directions. At the chill, crystals which have nucleated evolve their latent heat as they grow. Thus, as they grow parallel to the chill, they are growing into metal which is at a lower temperature. Chalmers$^{24}$ has explained why this should result in dendritic growth, even for pure metals. Dendrites which are more nearly oriented with their $\langle 100 \rangle$ axes in the plane of the chill should grow to a larger size, on the average, than grains of another orientation. Still, smaller grains would be expected of other orientations; in fact, for the case of random nucleation at the chill a preferred texture would result only because some grains grow larger than others. In their study of "The Origin of the Preferred Orientation in the Columnar Zone of Ingots", Walton and Chalmers$^{23}$ studied the change in preferred orientation in directionally solidified aluminum ingots with distance from the chill. Their results showed no preferred orientation at the mold wall. However, the X-ray technique they used measured the preferred orientation in terms of grain numbers, not grain areas; larger grains were weighed equally with small ones. The term "preferred orientation" in their experimental work means "cluster preferred orientation". No "cluster preferred orientation" is expected for random nucleation at the chill, no matter what growth anisotropy exists. (Walton and Chalmers observed a sharp development of "absolute preferred orientation" in columnar grains under some conditions.) It is worth noting at this point that Edmunds' determination of preferred orientation at the
chill in the hexagonal metals took account of grain area, and is therefore an "area base preferred orientation".

In a later paper, Edmunds\textsuperscript{20b} stated that experiments showed that "Aluminum . . . die castings . . . have random grain orientation textures at the surface". No experimental details were given in the paper, except that the same method of orientation determination was used with aluminum as in the earlier paper on hexagonal metals.

3. Chill Grain Structure

To the best of the author's knowledge no work has been published on the dendritic structure of chill grains. (Biloni and Chalmers\textsuperscript{25} will publish soon an investigation of the "predendritic" structure of chill surfaces.) It is worthwhile, therefore, to discuss what might be expected. In the discussion of preferred orientation, it was suggested that the $\langle 100 \rangle$ directions should grow more quickly than the other directions. In this case we would expect grains with a $\langle 100 \rangle$ direction in the chill plane to be elongated in that direction, on the average.

Several investigators since 1929\textsuperscript{12,26-29} have noted that dendrite traces in a plane of polish are the intersection of $\{100\}$ planes with the plane of polish; that is, the morphology of fully solid dendrite growth forms is largely plate-like. This fact should also be reflected in the dendritic structure of the chill surface. If growth is more rapid in the $\{100\}$ planes than in the other planes, chill grain elongation along the chill surface might be expected in the direction of the intersection of a $\{100\}$ plane with the chill surface.
The three \(\{100\}\) planes in a cubic system are, of course, mutually perpendicular. However, if they are intersected by a random plane (hkl) and viewed in two dimensions, as occurs in the observation of dendrites, non-orthogonal traces may be seen. This has already been studied in columnar grains\(^{27,29}\) and is discussed in Chapter III. Chill grains, however, are more random than columnar grains, so it is useful to show the wide variation possible in the two dimensional aspect of the intersection of \(\{100\}\) planes with a plane (hkl). The angle between two traces representing two intersections of \(\{100\}\) planes with the plane under consideration (the chill plane) may vary from 90° for one pair of intersections in all grains oriented along the \(\langle100\rangle\) zone, to 60° for all three pairs of intersections in the case of a grain of \(\langle111\rangle\) orientation (see Appendix A for the analysis).

Figure II-1 shows the angle of intersection of pairs of \(\{100\}\) planes versus orientation in the stereographic triangle, considering only the two \(\{100\}\) planes making the largest angle to the chill plane. In summary, then, if the dendrites are plate-like \(\{100\}\) planes, then the two-dimensional aspect of chill dendrites will be that of intersection of dendrite traces at angles between 60° and 90°.

B. Experimental Apparatus

In this investigation, a new technique was developed for the study of chill structures. It might be useful to summarize the difficulties encountered in the study of chill grains and how they were overcome.

1. Chill grains are subject to exudation.\(^{30}\) This is a layer of high solute liquid which runs out of a casting during the late stages of solidification. It covers the original surface of the casting (the chill
Figure II-1: The angle between traces formed by the intersection of \( \{100\} \) planes with various other planes \( \{hkl\} \). The \( \{hkl\} \) planes are represented by dots in the stereographic triangle. Only the \( \{100\} \) planes making the two highest angles to the plane \( (hkl) \) are considered in each case. The plane \( (hkl) \) represents the crystallographic plane of the chill grain which is parallel to the chill surface (i.e., the orientation of the chill grain).
surface) with a layer of solute enriched solid. In order to study the chill layer, the exuded layer must be removed without damaging the chill structure. Better still, it should be avoided.

2. Chill dendrites have a very fine structure and are sometimes difficult to etch.

3. Chill structures tend to reflect mold structures quite closely. Any defects in the mold are reproduced in the chill structure. Perfect reproduction is prevented in part by the interdendritic shrinkage which occurs.

4. Chill surfaces are subject to defects known to foundrymen as scabs, cold shuts, and blows. Scabs result from liquid splashing onto the chill and solidifying before the main body of liquid arrives; liquid then freezes on the scab without fusing to it. Cold shuts are caused by flowing metal from two independent sources failing to flow together completely to fill the mold. Blows are due to either trapped air between the mold and the metal or to water on the mold, both of which expand in contact with hot metal.

The apparatus which was designed overcame each of these difficulties and made use of one of them. A polished copper chill was used and interdendritic shrinkage caused the first-formed dendrite projections to stand in relief. See Figures II-3a - h.

An amorphous carbon mold coating was used to increase fluidity and allow the casting to be filled without cold shuts. The metal was drawn up into the mold by a partial vacuum of 4 centimeters of mercury. See
Figure II-2. This allowed the mold to be filled quickly and smoothly without scabs or blows. Several defects remained, all of a minor nature. Some pits appeared on the surface, probably due to trapped gas. Also, a dark film obscured parts of the chill surface in some cases, especially at higher casting temperatures. This was no doubt an oxide film, most of which must have formed while the mold was filling. Several castings had hot tears due to hindered contraction which causes intergranular fracture.32 Finally, in some cases very little or no interdendritic shrinkage occurred at the chill face, so the structure was not revealed.

C. Mold Design

Castings were made of varying thickness and height and with several pouring temperatures. The thickness of the casting was determined by the thickness of the shims used (see Figure II-2); shims were 1/25 inch, 1/8 inch, and 3/8 inch thick. The sides of the mold were identical polished copper blocks of two sizes. One size was 3 inches wide, 3 inches high and 1 inch thick each, while the other size was 3 inches by 1 inch by 5 inches high. The shim width on each side was 1/2 inch, so the resulting castings were 2 inches by 3 inches or 2 inches by 5 inches with varying thickness (unless the mold did not fill completely). Steel shims used in the first castings were replaced with Fiberfrax coated transite shims to prevent edge effects. Melting practice is discussed in Appendix B.

Castings discussed in this thesis are listed in Table II-1.
Figure II-2: Apparatus used for making chill dendrites. Liquid metal was drawn up into a mold consisting of two polished copper blocks, two shims of variable width, a top, and four nuts and bolts. A steel pipe was threaded into the top.
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<td>870</td>
<td>1/25</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>870</td>
<td>3/8</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>730</td>
<td>3/8</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>790</td>
<td>1/25</td>
<td>3</td>
<td>titanium refined</td>
</tr>
</tbody>
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D. Results

1. Grain Size.

Effect of Pouring Temperature and Section Size.

Figures II-3 and II-4 show surface structures on castings listed in Table II-1. Three effects are apparent in these castings. Grain size generally increases with increasing pouring temperature, although the
Figure II-3: Chill castings. (a) Casting 1 (700°C, 3/8-inch thick) 
(b) Casting 2 (790°C, 3/8-inch thick) (c) Casting 4 (790°C, 
3/8-inch thick) (d) Casting 7 (870°C, 1/8-inch thick) 
Steel shims were used in these castings.
Figure II-3 (continued):  
(e) Casting 5 (700°C, 1/8-inch thick)  
(f) Casting 8 (700°C, 1/25-inch thick)  
(g) Casting 9 (700°C, 1/25-inch thick)  
(h) Casting 12 (870°C, 1/25-inch thick)  
Transite shims were used in these castings.
Figure II-4a: Casting 6 (760°C, 1/8 inch thick). One micron alumina spray coating used instead of amorphous carbon coating.
Figure II-4b: Casting 10 (780°C, 1.25 inch thick).
effect is not a large one; the largest grains in the 1/25 inch and 1/8 inch plates poured at 700°C are about the same size as the largest ones in the same plates poured at 870°C. These grains may be more than one centimeter in diameter. In the 3/8 inch thick plates a higher pouring temperature seems to give larger grains (except that castings poured at 870°C have a thick oxide film and the grain size is no larger than at 790°C). Some grains are four or more centimeters in one dimension. A second effect is the change in grain size with plate thickness. Thicker plates have a somewhat larger grain size than thinner ones at a given pouring temperature.

In the preceding paragraph only the larger grain size was discussed in castings where a duplex grain size existed (Figure II-3, f and g). The existence of this duplex size is the most outstanding grain size effect in the chill casting series. It is significant to note that the fine grains associated with a duplex structure were always found in castings that failed to fill the mold. At the same time, large grains were also present, usually toward the bottom part of these castings. An explanation of this duplex structure is given in Appendix C. Basically, it seems that the convection associated with filling the mold refines the grain size of the solidifying metal. The large grains are found in the lower part of the casting where solidification occurs after flow has stopped. The fine grains are typically one or two orders of magnitude smaller than the large grains, about .2 millimeters in diameter.
2. **Chill Dendrite Structure**
   
   a. **Dendrite Trace Geometry.**

   Figure II-5 is an enlarged photograph of the chill surface of Casting 13, which was drawn into a chill mold at 730°C. Each surface grain has a different dendritic pattern, although there are some parts which are less clear than others. The orientation of each grain was determined by the Laue back reflection method. In some cases, several orientation determinations were made on the same grain. For example, separate checks were made on different regions to ensure they were indeed part of the same grain. In other cases, what seemed to be separate "grains" actually had almost the same orientation. In cases where several determinations were made and the orientation was similar enough in two regions to be considered one grain, but showed some differences, the grain was divided into parts according to appearance (A, B, etc.), although this does not mean that the orientation was constant in these areas. A diagram of the grains of Casting 13 and the numbers assigned to those grains is given in Figure II-6. Figure II-7 is a stereographic triangle showing the orientation of grains in Casting 13.

   A cursory examination of Figure II-5 shows that, in any single grain, the traces of one set of dendrite axes are roughly perpendicular to the trace of a second set of axes. This is in contrast to the angles (shown in Figure II-1) that would be expected if only \{100\} dendrite sheets developed, and these sheets intersected the chill surface. An explanation follows using specific grains as examples.
Figure II-5: Casting 13 (730°C, 3/8 inch thick). The grains of this casting are outlined in Figure II-6. The orientation of all numbered grains was determined by the Laue back reflection X-ray technique.
Figure II-6: Grains of Casting 13, showing the numbering system used and the Laue X-ray spot location. Dots indicate orientations given in Figure II-7. Crosses indicate other orientation determinations.
Figure 11-7: The orientation of grains of Casting 13. The variation of dendrite trace geometry from orthogonal to non-orthogonal is indicated.
Grain 65 can be found in the upper center of Figure II-5; a higher magnification photograph is shown in Figure II-8. The Laue photographs of this grain gave sharp undistorted spots. All Laue photographs were made with the casting aligned vertically, as shown in Figure II-5, so a correlation can be made between the surface structure and the X-ray analysis. The stereographic projection of Grain 65 is shown in Figure II-8. Great circles have been drawn through the \(\{100\}\) zones in the projection; each great circle intersects the circumference of the stereographic projection at two points \(180^\circ\) apart. A straight line joining these points (which also passes through the center of the stereographic projection) shows the orientation of the intersection of a \(\{100\}\) plane with the plane of the page. If the dendrite traces of Grain 65 are caused by the intersection of \(\{100\}\) dendritic planes, then the orientation of the line in the stereographic projection will be the same as the orientation of the trace in the chill surface. Examination shows that both sets of the orthogonal traces visible in Grain 65 correspond to the orientation of the intersection of \(\{100\}\) planes, one of which is oriented almost perpendicular to the chill surface. Therefore, the structure of this grain corresponds to the predictions made in the introduction to this chapter.

Past studies have shown that the \(\langle 100 \rangle\) direction is important in dendritic growth. This is supported by the results typified by Grain 17, which may be found in the upper left of Figure II-5, shown at higher magnification in Figure II-9. Again, orthogonal dendrite traces are apparent in this photograph. A comparison with the stereographic projection, however, shows that only one set of traces has the orientation of the intersection of \(\{100\}\) planes. A line drawn through the stereographic projection
Figure II-8: Stereographic projection of the Laue photograph of Grain 65 with the microstructure in a corresponding orientation. The dendrite traces of this grain, which lies on the ⟨100⟩ zone, are at 90° as predicted in Figure II-1.
Figure II-9: Stereographic projection and corresponding photomicrograph of Grain 17. This grain is oriented between $\langle 111 \rangle$ and $\langle 100 \rangle$, as shown. Of the orthogonal dendrite traces, one set corresponds to intersections of $\langle 100 \rangle$ planes with the chill surface, the other set to projections onto the chill surface of $\langle 100 \rangle$ directions of like indices. A second set of $\langle 100 \rangle$ planes (intersecting the chill $59^\circ$ from perpendicular) makes non-orthogonal traces on the chill surface.
parallel to the second set of traces passes through a $\langle 100 \rangle$ pole, as shown in the figure. Thus, this set of traces has the orientation of the projection of a $\langle 100 \rangle$ direction onto the chill surface. The intersection of a $\{100\}$ plane with a surface $(hkl)$ will always produce a line which is perpendicular to the projection onto the plane $(hkl)$ of a $\langle 100 \rangle$ direction having identical indices as the $\{100\}$ plane. Therefore, all chill dendrites having intersections of a $\{100\}$ plane in combination with projections of a like-indexed $\langle 100 \rangle$ direction will show orthogonal surface traces. In the case of Grain 65 projections of $\langle 100 \rangle$ directions coincide with intersections of $\{100\}$ planes. A third set of traces is visible in Grain 17. This set corresponds to intersections of $\{100\}$ dendrite sheets oriented at $59^\circ$ from perpendicularity to the chill surface. The orthogonal dendrite plane intersection was at $22^\circ$.

Grain 60 oriented with a $\langle 111 \rangle$ direction nearly perpendicular to the chill surface is shown in Figure II-10. It is located at the lower left of center in Figure II-5. Some scratches are apparent in the picture; these scratches were the result of handling during the X-ray examination of the casting. The dendritic structure is still clearly apparent, however. Orthogonal dendrite traces are not the most obvious feature of the structure. Dendrite traces seem to branch in every direction. Actually, intersections of all three $\{100\}$ planes and projections of two of the three $\langle 100 \rangle$ directions may be identified by comparison to the stereographic projection. Grains 8A and 42C are two other grains having orientations near the $\langle 111 \rangle$ pole of the stereographic triangle. These are shown in Figures II-11a and b. Again, their traces are not obviously orthogonal due to the development of traces representing the intersection of all three $\{100\}$ planes.
Figure II-10: Stereographic projection and corresponding photomicrograph of Grain 60. A $\langle 111 \rangle$ direction of this grain is almost perpendicular to the chill surface. Traces representing the intersection of all three $\langle 100 \rangle$ planes can be seen in the photomicrograph. Also, one projection of a $\langle 100 \rangle$ direction is strongly developed (sloping from left to right, almost horizontal in the picture).
Figure II-11: Other grains oriented with a \(<111>\) direction nearly perpendicular to the chill surface. These also show non-orthogonal dendrite traces.
The characteristic angle between traces for grains near the $\langle 111 \rangle$ pole is in agreement with the calculations of Figure II-1, while grains near the $\langle 100 \rangle$ zone show an orthogonal structure. A careful study of all grains of Casting 13 shows that there is a region near the $\langle 111 \rangle$ pole where the chill structure appears to be non-orthogonal due to the growth of several $\{ 100 \}$ planes. Nearly all other grains give a basically orthogonal appearance. The approximate zone separating these two regions of the stereographic triangle is shown in Figure II-7.

In most cases the projection of $\langle 100 \rangle$ direction or intersection of $\{ 100 \}$ plane which is represented by a developed dendrite trace in the chill surface is the direction making the smallest angle with the chill surface or the plane most nearly perpendicular to the chill surface. In the case of like-indexed (orthogonal) projections and intersections, these angles are identical. Therefore, non-orthogonal dendrite traces represent development of a second lowest angle $\langle 100 \rangle$ projection or (more usually) $\{ 100 \}$ intersection (e.g., Grain 17). One grain was found in which the second lowest plane and direction formed orthogonal traces. In several other grains the second lowest angle plane or direction combined with the lowest angle direction or plane respectively to give a non-orthogonal structure, usually with an angle not far from $90^\circ$.

It is of interest to find the relationship between surface chill structure and structure within the casting. As shown above, some traces on the chill surface are intersections of $\{ 100 \}$ planes with the chill surface. But does the $\{ 100 \}$ plane extend into the casting? Also, other dendritic traces on the chill surface are projections of $\langle 100 \rangle$ direction onto the chill
surface. Two cross-section structures are shown in Figure II-12. Section (a) is a section from Casting 3, 3/8 inch thick, poured at 760° C. Section (b) is a section from Casting 12, 3/8 inch thick, poured at 870° C. These two sections are typical of others which were examined. Several observations can be made. First of all, dendrite traces perpendicular to the chill surface are seldom evident near the chill surface. Thus, projections of \( \{100\} \) directions seen on the chill surface do not represent dendritic planes. Instead they must result from dendritic growth along the chill in a non-crystallographic direction represented geometrically by the projection of a \( \{100\} \) direction. In Figure II-12a a lower solute (light etching) layer is visible at the chill surface. This same structure is discussed in Chapter III and is clearer in the coarser structures presented there. This thin layer must be the low solute region formed by the growth of projections of \( \{100\} \) directions in the chill plane. Also in Figure II-12a some dendrite traces can be seen making an angle of about 30° to the chill surface. These are \( \{100\} \) dendrite planes growing from the chill. The traces at 90° to these, some of which grow right from the chill surface, are also \( \{100\} \) planes. In Figure II-12b the structure near the chill is quite different from that beyond .4 millimeters from the chill. Near the chill, dendrite traces are about equally well developed parallel and perpendicular to the chill. In this region, the dendrite probably grew along the chill surface. Beyond .4 millimeters (from the chill surface (2 centimeters at 50 magnification) the dendrite traces are extended roughly perpendicular to the chill. In this region, the grain grew away from the chill surface.
Figure 11-12: Cross-section structure at the chill surface (sectioned, polished, and etched). (a) Casting drawn at 760°C, 3/8 inch thick. A low solute layer exists at the chill surface. Also, dendrite planes at a low angle (30°) to the chill are obvious at the right (50X). (b) Casting drawn at 870°C, 3/8 inch thick. Within .4 mm of the chill surface, the structure appears to result from growth along the chill surface. Further from the chill, the dendrite sheets are elongated perpendicular to the chill (50X).
Thus, it seems that there are two mechanisms by which a grain grows along a chill surface. One mechanism is by columnar dendrite growth in a direction at a low angle to the chill surface. This results in dendrite planes lying at low angles to the chill surface. Comparison of the castings of Table II-I shows that this mechanism is more common at higher pouring temperatures and greater casting thicknesses. The second mechanism is growth of dendrite traces on the chill surface. This is discussed below.

Chalmers has explained that dendrites grow in a \langle100\rangle direction because this is the direction bounded by the four slow growing \{111\} planes. Since an actual dendrite tip does not seem to have well developed facets, the dendrite direction must be controlled by other planes as well. When growing on a chill surface, some dendrite axes grow in a direction represented by the projection of a \langle100\rangle direction. A possible explanation for this change in growth direction from the usual \langle100\rangle direction follows. The surface energy relationship between solid (dendrite tip), liquid melt, and the thin oxide layer adhering to both is assumed to result in a semi-dendrite tip, as shown in Figure II-13. The energy relationship is \( \gamma_{SO} = \gamma_{LO} + \gamma_{SL} \cos \theta \), where \( \gamma_{SO} \) is the solid-oxide interface energy, \( \gamma_{SL} \) is the solid-liquid interface energy, \( \gamma_{LO} \) is the liquid-oxide interface energy, and \( \theta \) is the contact angle. If \( \gamma_{LO} > \gamma_{SO} \), less than half of the dendrite tip will be present. In Figure II-13 the energy relationship was assumed to be such that a \langle100\rangle direction lies perpendicular to the dendrite tip at the chill interface. Since this is the fastest growing direction, growth will be most rapid at the chill interface, and the dendrite tip will propagate on the interface. At the same
Figure II-13: Sketch of a semi-dendrite tip growing on the chill surface as the projection of an (100) direction.

Figure II-14: Grain e of Casting 13.
1. Laue photograph (2 hour exposure).
2. Photomicrograph (50X).

Figure II-15: Laue X-ray beam size and diffracted spots from a non-dendrite (Grain 66).
1. Laue X-ray beam (50X).
2. Laue photograph (20 minute exposure).
time, the other dendrite surfaces present on the "semi-dendrite" tip keeps it from going left or right and hence hold it in the $<100>$ projection.

Growth away from the chill surface region occurs by dendritic growth of $\{100\}$ dendrite planes oriented according to the original nucleus orientation. The growth of $\{100\}$ dendrite planes and their intersection with the chill surface at relatively high angles must have the same explanation as the formation of dendrite planes in columnar grains (see Chapter III).

b. Macro-Mosaic Structure.

Grain 6 of Casting 13 is an interesting one. The first Laue photographs showed no pattern, after 10 minute exposures. (All other grains gave patterns with sharp spots after a 10 minute exposure.) A two hour exposure was made and is shown in Figure II-14a. Clusters of spots are visible in several locations. The average orientation can be determined from these clusters; this orientation corresponds to the macroscopic structure of Grain 6 shown in Figure II-5. The spots which make up the cluster represent small regions of the grain having a definite orientation. The grain therefore has a "macro-mosaic" structure described by Buergers.33

The origin of this structure is of interest. Examination of Figure II-5 shows that parts of Grain 6 show no clear dendrite traces. A higher magnification photograph (Figure II-14b) in the area of the Laue photograph shows two regions. In the lighter areas dendrite traces can be seen on the surface. In the darker areas only small light spots can be seen. It seems reasonable that dendrite traces visible on the chill surface result from growth on the chill surface. Discreet spots must result from growth
toward the chill surface. Independent dendrite elements would develop during growth toward a chill surface just as they do during columnar growth (see Chapter III) and misorientation could develop between them. Misorientation of several degrees have been reported in the literature for columnar growth. Four to six degree misorientations can be measured in any of the clusters of Grain 6.

Some of the spots in the cluster are sharper than others, probably because they correspond to a larger area of constant orientation. Figure II-15a shows the Laue spot size at fifty magnification, which can be compared directly to the structure of Grain 6 in Figure II-14b, and the resulting Laue pattern (Figure II-14a). The line of sharp spots probably corresponds to "secondary" dendrite traces along a relatively well developed "primary" trace. The multitude of fine spots, on the other hand, correspond to dendrite elements that grew toward the chill surface, shrinkage between those resulting in the darker areas on the photomicrograph.

c. Bent Dendrites.

Bent dendrite traces can be seen on the chill surface of most of the castings made (for example Casting 6, Figure II-4a). The radius of curvature varies from very large radii down to 1 - 2 centimeters (for larger grains; convection refined grains will be discussed later). Qualitative comparison of the castings indicates that more bending occurs the thinner the section thickness, and that dendrite bending is not dependent on pouring temperature. Since cooling rate depends on section thickness (see Appendix D) rather than pouring temperature, extent of dendrite bending may be a function of cooling rate.
X-Ray Analysis: Typically, a bent dendrite has a Laue pattern such as Figure II-15b, which is from Grain 60, an undistorted grain. The orientation change over distances the size of the X-ray spot are not large (less than 1/2°). Over larger distances, the orientation changes may be very large indeed; a 40° change in orientation is not unusual.

There are two components of the orientation change which may be resolved. One component is the bend which is observable in the chill surface; this is due to a rotation of the crystal lattice about an axis perpendicular to the chill plane. The second component is a change in orientation in the stereographic projection; this component is due to the rotation of the crystal lattice around an axis lying in the chill plane. All bent dendrites show a change in orientation in the stereographic projection. Also, one dendrite which was not bent in the chill plane showed a change of orientation in the stereographic projection. The dendrite trace which is bent may be either the projection of a $\langle 100 \rangle$ direction or the intersection of a $\{ 100 \}$ plane.

Consider the change of orientation in the stereographic projection along a bent dendrite. Two cases are possible (see Figure II-16). Part (a) shows the results that would be obtained if the dendrite crystal lattice were convex toward the chill. The change of orientation would occur in such a way that a movement along the bent dendrite from left to right, for example, would cause the stereographic projection to move in the same direction. Part (b), on the other hand, shows what would be expected in the case of a dendrite crystal lattice that was concave toward the chill. In this case, movement along the dendrite from left to right would cause a movement of the stereographic triangle from right to left. Part (c) shows the results
Figure II-16: Analysis of bent dendrite. A. Schematic diagram of crystal lattice with a bend convex to the chill surface and corresponding orientations at various points, indicated in stereographic triangles. Looking from left to right, the orientation dot moves from right to left in stereographic triangle. B. For a concave bend the orientation dot moves from left to right as the dendrite is examined from left to right. C. A bent dendrite from Casting 6 (3X) and portions of the stereographic projections at points 1, 2 and 3 on the bent dendrite trace. The bend of the dendrite corresponds to b; it is concave to the chill surface. The bend observable in the chill surface is reflected in a corresponding rotation of the stereographic projection from 1 to 2 to 3.
obtained on an actual bent dendrite from Casting 6 (Figure X). The result was the same for all (large grain) bent dendrites, and is qualitatively to Figure II-16b. That is, all bent dendrites are bent to the chill surface.

Bending of a dendrite trace is associated with a change of the grains in the stereographic projection. An associated two rotations indicates that bending occurs about an axis at a between 0° and 90° to the chill surface (and is observed results in two components). It seems probable that thermal gradients are a dendritic plane, and that this is resolved into a bend of the chill surface and a change in orientation. Bending of a projection 〈100〉 direction must result from bending of a dendritic plane associated with the projection of direction which bends. And the thermal gradients necessary to cause dendrite bending, with those which exist under the conditions when bent dendrite is given in Appendix D.

d. Convected Grain Structure.

Fine grains were found in some castings, such as 8, 9 and 10, failed to fill the mold. It was suggested above that these forms while the metal was filling the mold. The structure will now be discussed. Figure II-17a shows the structure of Casting 8. Both light and dark areas can be seen, as in Grain 13. Some bending of the dendrite traces is also visible. A photograph of the structure is shown in Figure II-17b. Several grains simultaneously exposed. It is obvious however, that a macro-
Figure II-17: Photomicrograph and BSE image of a fragment of the fine grains resulting from movement of Casting F. (a) Surface structure of fine grains in Casting F. Light areas are dendrite traces that remain on the chill surface. Darker (spotted) regions are areas where dendrite growth forms grew toward the chill surface and surface left a rough surface (50X). (b) Late portion of region in above structure. Late spot size as in Figure II-16b. Clusters of spots indicating substructure within a dendrite can be seen. Also, an arc of strong spots at 2:00 o'clock probably results from a light region where dendrite elements would have a large area.
structure like that of Grain 6 exists. Clusters of spots exist in several areas. There are several lines of bright spots, probably due to reflections from bent dendrite traces that grew on the chill. Others are more diffuse spots, caused by reflections from the dark areas on the photomicrograph; these are dendrite elements that grew toward the chill. Figure II-18 shows a cross-section of Casting 8. Section (a) is taken from the coarse-grained region of the casting. Notice that one grain extends across the thickness of the casting, indicating interface heat transfer control (see Appendix D). Section (b) shows the sharp change in structure from coarse to fine grains. The grains do not appear severely bent in cross section, indicating that surface traces may be more severely bent than the internal structure. Section (c) shows large grains that grew at the top of the flowing stream. Here convection would be less severe and fewer grains were nucleated.

Figure II-19 shows some of the severely distorted surface structure found in this casting. These photographs are at 50 magnifications, so the dendrite bending is far more severe than in the bent dendrites discussed previously. Radii of curvature as low as 1 millimeter can be seen. Perhaps convection greatly increases surface temperature gradients which cause this severe and often very non-uniform bending.

e. Grain Refined Structures.

Several castings were intentionally grain refined by additions of titanium. Figure II-20a shows the structure of Casting 14 drawn at 790°C into a 1/25 inch thick mold. Compare Casting 8 (Figure II-17a) drawn at 700°C into the same mold. The general appearance is similar. No dendrite bending is obvious in the titanium grain refined structure, however.
Figure II-18: Structure perpendicular to the chill in Casting 8 (700°C, 1/25 inch thick). (Magnification, 50X)
(a) Lower part of the casting showing coarse grains, some of which extend across the section.
(b) Middle of the casting, showing the transition from coarse grains to convection refined grains.
(c) Top of the casting, showing several coarse grains.
Figure II-19: Severely distorted surface structures in convected surface drains. The radius of curvature of bend observable in the mill surface may be 1.5 cm. or less. (Magnification 50X)
Figure II-20: Titanium grain refined Casting 14 (790°C, 1/25 inch thick). (a) Chill surface structure. The structure is similar to that of convection refined grains, except that no bending is evident (magnification 50X). (b) Laue photograph, showing clusters of spots but not lines of spots, indicating that misorientation is due to growth toward the chill rather than on the chill. (c) Electropolished chill surface (magnification 50X). Electropolishing produces the same structural appearance in the light and dark regions of the unpolished surface.
Figure II-20b shows a Laue photograph of this structure. Again, clusters of spots are evident, corresponding to the dark regions of Figure II-20a, where dendrite elements grew toward the chill. No lines of spots of high intensity, however, due to bent surface dendrite traces, are found in the photograph (compared Figure II-17b).

3. **Preferred Orientation**

Measurements of preferred orientation on the surface of grain refined castings by the inverse pole figure method showed no definite preferred orientation. This might be attributed to the small grain size. That is, if the dendrite did not develop fully, preferred orientation would not be expected. However, the dendrites were quite well developed in these fine grains, as shown in Figure II-20c.

In part 2 of the introduction to this chapter, expectations about the development of preferred orientation were discussed. If \( \langle 100 \rangle \) directions grow significantly faster than others, a grain oriented with \( \langle 100 \rangle \) directions parallel to the chill surface should grow larger than other grains. A \( \langle 100 \rangle \) grain has two \( \langle 100 \rangle \) directions parallel to the chill surface. A grain on the \( \langle 100 \rangle \) zone has one \( \langle 100 \rangle \) direction parallel to the chill surface. More rapid growth of \( \langle 100 \rangle \) directions or \( \{100\} \) planes should both tend to most favor a \( \langle 100 \rangle \) orientation, and to give some preference to grains on the \( \langle 100 \rangle \) zone (having one \( \langle 100 \rangle \) direction parallel to the chill and one \( \{100\} \) plane perpendicular to the chill). The extent of preference is measured below both as a function of angle from the \( \langle 100 \rangle \) pole and as a function of angle from the \( \langle 100 \rangle \) zone.
In Figure II-7 circles are drawn in the stereographic triangle at 5° intervals from the \( \langle 100 \rangle \) pole. For a random grain orientation, the pole of the grain would be just as likely to be in one orientation as any other; that is, the poles would be spread evenly over the surface of the reference sphere. Therefore, the number of grains in any angle range from the \( \langle 100 \rangle \) direction would be proportional to the area on the reference sphere in that angle range. The calculation of the areas in 5° angle ranges from the \( \langle 100 \rangle \) pole is given in Appendix E. These areas are shown in Figure II-21a, a bar graph showing expected number of grains versus angle range from the \( \langle 100 \rangle \) pole for a random grain orientation.

Figure II-7 also shows lines drawn at 10° intervals from the \( \langle 100 \rangle \) zone (the line joining the \( \langle 100 \rangle \) and \( \langle 110 \rangle \) poles in the stereographic triangle); these lines are latitude lines of the Wulff net for a standard \( \langle 100 \rangle \) stereographic projection. A calculation of the areas in 5° angle ranges from the \( \langle 100 \rangle \) zone is given in Appendix E. These areas are shown in Figure II-21c and represent the expected number of grains versus angle range from the \( \langle 100 \rangle \) zone for a random grain orientation.

Figure II-7 gives the orientation of surface grains in Casting 13. When plotted as bar graphs like those of Figure II-21a and II-21c, the results can be compared to random grain orientation. This is done in Figures II-21b and II-21d. Figure II-21b shows a slight deficiency of grains for 0° - 5° from the \( \langle 100 \rangle \) zone and Figure II-21d shows less than a random number of grains from 0° - 15° from the \( \langle 100 \rangle \) pole. Overall, however, the distribution of orientations is very like a random distribution, and differences may be the result of a small sample size.
Figure II-21: Grain orientation distribution on the chill surface. 
(a) Random grain distribution in $5^\circ$ increments from the $\langle 100 \rangle$ direction (theoretical). 
(b) Number of grains in Casting 13 versus angle range from the $\langle 100 \rangle$ direction. 
(c) Random grain distribution in $5^\circ$ increments from the $\langle 100 \rangle$ zone (theoretical). 
(d) Number of grains in Casting 13 versus angle range from the $\langle 100 \rangle$ zone.
This is consistent with Walton and Chalmers' results, and is expected if random nucleation occurs.

If there is a preferred orientation, it must result from a variation of grain size with orientation. The areas of grains shown in Figure II-6 were measured. Average grain size versus angle range from the \(<100\) pole is shown in Figure II-22a. The range \(0^\circ - 20^\circ\) is shown as one bar due to the small sample in that range. The average grain size seems larger in the range \(0^\circ - 35^\circ\) than \(35^\circ - 55^\circ\), but the effect (if real) is not large. Average grain size versus angle range from the \(<100\) zone is shown in Figure II-22b. Here, the \(25^\circ - 35^\circ\) range is shown by one bar, since this is also a small sample. The result is clear; very little difference in average grain size exists.

Thus, the expectations that favorably oriented grains might grow much larger than others at the chill is not borne out by experiment. Instead, the results indicate that there is no major difference in growth rates of grains at a chill surface as a function of their orientation. Small differences in growth rates are not ruled out, however. The mechanism of the development of preferred orientation in columnar grains depends on the competition of one grain against another. In the case of growth at a chill surface, small differences in growth rate will produce only the same small differences in average grain size. In columnar growth, small differences of growth rate or any other factor are amplified by competition into a sharp preferred orientation.
Figure II-22: Average grain size versus orientation in Casting 13.
(a) Grain size versus angle range from the $\langle 100 \rangle$ direction.
(b) Grain size versus angle range from the $\langle 100 \rangle$ zone.
E. Summary

A new technique has been described for studying the surface structure of alloys cast against a chill. The technique involves solidification of a thin section casting in a smoothly polished metal mold. Typical surface structures produced in aluminum-4.5 per cent copper alloy are shown in Figures II-3 and II-4.

The surface grain size depends strongly on the casting conditions. When solidification proceeds while the metal is filling the mold, fine grains result (Figures II-4b and II-17). When the mold fills completely before the start of solidification surface grains 100 times as large (3 centimeters or more in one dimension) may be observed (Figure II-4a). Convection during solidification appears to be responsible for the fine grain size observed in the former case.

The dendrite "arms" visible on the chill surface usually are orthogonal in a single dendrite (see Figure II-7). One set of orthogonal dendrite traces ("arms") consists of the projections of $\langle 100 \rangle$ directions onto the chill surface; the orthogonal set of "arms" consist of intersections of $\{ 100 \}$ planes with the chill surface. The orthogonal planes and directions are like-indexed.

When a $\langle 111 \rangle$ direction in the dendrite lies nearly perpendicular to the chill surface, the dendrite arms do not appear orthogonal (Figure II-11). In this case, two or three $\{ 100 \}$ plane intersect the chill surface and form dendrite traces. (The projections of one, two or three $\langle 100 \rangle$ directions may also be present.)
Laue X-ray analysis of a large number of chill grains shows that their orientation is random (Figure II-6). Also, the average grain size does not vary significantly for different orientations (Figure II-22).

Dendrite bending occurs under some casting conditions. Moderate bending of dendrite traces can be seen in some large surface grains (e.g., Figure II-4a). This bending is concave toward the chill surface (Figure II-16). The orientation change in these dendrites may be large from one end of the grain to the other, but a Laue photograph at any point is sharp. Bending in fine grains is much more severe, if these are produced in castings which solidify while the mold is filling, under strong convection (see Figure II-19). If fine grains are produced with a grain refiner but with no convection, no bending is apparent (see Figure II-20). The radius of curvature of bend in large grains is ten or more times the radius observed in fine grains (in the chill surface).

Several conclusions based on the results of this Chapter may be made:

(1) Chill surface structures may be studied using a polished chill. Shrinkage between dendrite "arms" reveals the structure.

(2) One way to study solidification under conditions of very low convection is apparent from the results. If the metal is drawn up by a partial vacuum into a thin mold before it begins to freeze, little convection will occur during solidification.

(3) The fine "chill zone" often observed in ingot making would seem not to result from the rapid chill, but from convection during pouring.
(4) Since a preferred orientation does not seem to develop at the chill surface (at least in aluminum alloys) other ways for controlling orientation in cast alloys must be sought.

(5) The dislocations introduced by dendrite bending at the chill surface might be studied. This study could lead to a better understanding of the origin of dislocations during solidification.
III. COLUMNAR DENDRITE STRUCTURE

A. Introduction
   1. General.

   Columnar dendrites comprise the second zone of a typical ingot. They originate in the chill zone and sometimes are terminated by an equiaxed zone. Columnar dendrites are grains characterized by having a dimension in the heat flow direction which is much larger than their dimensions perpendicular to heat flow. The difference between columnar dendrites and chill dendrites is one of degree. Columnar dendrites are those chill dendrites which grow into the ingot from the chill surface; other chill dendrites are suppressed.

   Various investigations are of interest in the understanding of columnar dendrites. These range from studies of massive ingot structure to laboratory investigations of small ingots and single crystals, and theoretical work on "constitutional supercooling" and instability of growth. In some cases understanding of columnar crystals parallels understanding of chill and equiaxed grains. In other cases attention is focused on certain aspects of columnar grains themselves: the development of preferred orientation, change in grain size, growth direction, and the morphology itself. This chapter is concerned primarily with the morphology of columnar dendrites.
2. Preferred Orientation and Structure.

Both phenomenological and theoretical studies have been made on the preferred orientation and structure of columnar grains, sometimes relating to dendritic growth generally. Nix and Schmidt\textsuperscript{5} studied the development of preferred orientation in columnar grains and noted that certain crystallographic direction become preferentially aligned with heat flow. Wineberg and Chalmers\textsuperscript{22,23} studied the growth of seeded dendrites of pure metal and found that the dendrite axes are extended in a low index crystallographic direction. Northcott and Thomas\textsuperscript{27}, in their study of columnar dendrites, noted that dendrite traces in a plane of polish represent the intersection of \( \{100\} \) planes with the plane of polish. Northcott's work confirms the work of Hultgren, mentioned in Chapter I.

Chalmers and co-workers have contributed greatly to the understanding of dendritic growth generally, with both their theoretical and experimental work. Chalmers' theory of constitutional supercooling is so well recognized that it need not be discussed here.\textsuperscript{24} Chalmers and Jackson\textsuperscript{24,36} have discussed the atomic kinetics of solidification in metals and alloys. Chalmers\textsuperscript{24} has explained the \( \langle 100 \rangle \) growth direction in dendrites as the direction bounded by four slow growing \( \{111\} \) planes. Walton and Chalmers\textsuperscript{23} have studied the development of preferred orientation in columnar growth. As mentioned in Chapter II, they noted the development of an absolute preferred orientation; this developed more quickly for higher alloy content and lower pouring temperature. Recently, Chalmers has summarized his and other work on "The Structure of Cast Metals" in his book, "Principles of Solidification".
3. Misorientation.

Misorientation develops during columnar growth between different parts of the same grain. Hultgren noted "twisting" of steel dendrite crosses in 1929. He also noted high cementite boundaries between dendrite crosses. Northcott observed a similar effect in aluminum in 1946. Winegard and Chalmers reported misorientation in dendritic single crystals of lead, zinc and tin of up to 30° between neighboring stalks. Striations and lineage observed in pure metals may be related to misorientation in dendrites. Striations are misorientation boundaries; they have been studied by Atwater and Chalmers and by Doherty and Chalmers. The results of Chapter II on chill grains may be added to the survey of misorientations which have been reported.

Doherty and Chalmers also studied dendritic growth in high purity aluminum. They observed both high angle (2 or 30°) boundaries like those noted above and also "sub-boundaries" which had a misorientation of less than 15 seconds of arc, the resolution of the technique used to measure misorientation in their experimental work. They explained the sub-boundaries as regions where two parts of the crystal having almost identical orientation grew together and formed a dislocation network. It is almost impossible for two parts of a crystal to grow together without forming a boundary. Therefore, dendrites, with their involuted structure, are an excellent source of boundaries, even in "pure" metals.

Many explanations have been proposed by the various investigators to explain the observed misorientations. Hultgren suggested that convection in the liquid is responsible. Weinberg and Chalmers suggest
that mechanical vibration of dendrites could be the cause. Rutter and
Chalmers have suggested that misorientations could be introduced in
cells when the lattice parameter must change to accomodate impurities.
Doherty and Chalmers consider that dislocations may be introduced
into the metal by collapsing vacancy disks. These are capable of
forming due to the excess vacancies caused by frozen in stacking faults.
Proheszka has suggested recently that thermal stresses due to
temperature gradients result in misoriented dendrite growth forms. When
these grow together, the observed misorientations result.

4. Growth Form Spacing.

Dendrites are the result of branching of dendrite growth forms. In
their paper (1953) Tiller et al used Chalmers theory of constitutional
supercooling to explain the instability of a plane front during growth,
the onset of branching. Sekerkta has recently refined this analysis.
These analyses apply only to condition for the breakdown of a plane
front, not to the resulting growth form spacing. The plane front con-
sidered may be a thickening dendrite growth form. Brown and Adams
have made an analysis to explain the observed spacing of high order
growth forms in a casting, which conform to the condition of constant
maximum constitutional supercooling. The detailed mechanism of growth
form spacing was not considered. The relationship Brown and Adams
obtained predicted that growth form spacing is proportional to the square
root of solidification time; the implication of the assumptions used is
that the growth form spacing considered was the minimum spacing in the
dendrite.
Experimental results on dendrite growth spacing have been variable. Several investigators have found that "dendrite arm spacing" is proportional to the square root of solidification time. Bardes has plotted data from several investigations on aluminum-4.5 per cent copper alloy over solidification times of $t = 2$ seconds to $4 \times 10^6$ seconds and found that minimum dendrite spacing is proportional to the $0.39$ power of solidification time. These data included results of Horwath and Mondolfo who found that minimum spacing was proportional to the fourth root of solidification time.

B. Experimental Method

1. Apparatus.

The apparatus used in this phase of the investigation has been described previously, and is shown in Figure III-1. The apparatus consisted of a recirculating air resistance furnace, a mold with bottom chill, and controlling and recording equipment. The furnace was used to maintain ambient temperature at $700^\circ$C, which is above the liquidus temperature of the aluminum-4.5 per cent copper alloy used. The cylindrical mold (5 inches diameter by 12 inches high) was insulated inside and outside, so that heat flow would proceed unidirectionally to the chill at the bottom. Water and compressed air were used as coolant to the chill. Thermal data was obtained from thermocouples placed in the ingot at various distances from the chill.
Figure III-1: Schematic diagram of the apparatus used in making columnar ingots. Coolants used were compressed air and cold water.
2. Procedure.

The mold and thermocouples were prepared and placed in the furnace; then the furnace was preheated to 700°C. About twenty pounds of master alloy was melted as described previously, and poured into the mold through a preheated type 304 stainless steel funnel (which was then removed). Chilling started within a few minutes, with compressed air or cold water circulated through the pipes and chill.

3. Castings Made.

Table III-1 indicates the coolant used in the ingots which are discussed in this chapter.

<table>
<thead>
<tr>
<th>Columnar Ingots Studied</th>
</tr>
</thead>
<tbody>
<tr>
<td>Columnar Ingot 1</td>
</tr>
<tr>
<td>2</td>
</tr>
<tr>
<td>3</td>
</tr>
<tr>
<td>4</td>
</tr>
</tbody>
</table>

* Ingot 11 of reference 29.

C. Results

1. Macrostructure.

Columnar Ingots 1, 2 and 3 were cut in half, polished and etched. Etching technique is described in Appendix E. The macrostructures of these ingots are shown in Figure III-2.
2. Chill Structure.

Microstructures of Ingots 1 and 3 at the chill surface and perpendicular to heat flow are shown in Figures III-4 and III-3, respectively. The structures are similar to cross sections at the chill in the chill plate castings of Chapter III, except that they are coarser. This is particularly true of Ingot 3 (Figure III-3) where four zones may be distinguished. The first zone, labelled "1", is a layer of high solute formed by inverse segregation (exudation) late in the solidification process. The second zone, labelled "2", is a layer of low solute at the original chill interface. The same structure, on a finer scale, is visible in Figure III-4, and to some extent in Figure II-12a. This layer must result in part from the growth of projections of \( <100> \) dendrite directions on the chill surface, as discussed in Chapter II. Dendrite sheets making a high angle to the chill surface will also contribute to the low solute region when they intersect the chill; they too grow along the chill. The third "zone", labelled "3" in Figures III-3 and III-4, results from the growth of \( \sum \frac{100}{2} \) dendrite sheets which lie at a low angle to the chill surface. This structure could be considered to be columnar growth along the chill surface. Zones two and three reflect the two ways that a grain grows perpendicular to macroscopic heat flow in the chill zone region. The last zone, labelled "4" in Figures III-3 and III-4, is typical of dendritic structures produced in unidirectional freezing.

This is the subject of discussion in the following sections. Thus, chill grains (columnar grains at the chill interface) may contain four structural zones, all within the same dendrite, but reflecting different stages in growth.
Figure III-3: Cross section at the chill surface of Columnar Ingot 3, showing four zones: (1) an exuded layer of high copper concentration, (2) a low solute layer corresponding to dendrite growth at the original chill surface, (3) dendrite sheets at low angles to the chill surface formed by columnar growth approximately parallel to the chill, and (4) columnar growth of orthogonal dendrite sheets at high angle to the chill surface (composite photograph, magnification 12X).
Figure III-4: Cross section at the chill of Columnar Ingot 1, showing zones 2, 3 and 4:
(2) low solute region at chill surface, corresponding to growth of dendrite growth
forms on the surface, (3) dendrite traces due to sheets at low angles to the chill
and (4) dendrite traces typical of growth away from the chill, caused by orthogonal
dendrite sheets at high angles to the chill (composite photograph, magnification 12X).

a. Water Chilled Ingot

Transverse sections (perpendicular to heat flow) from Ingot 1 are shown in Figure III-5. These sections are taken at 1/2 inch intervals from the chill surface. Larger sections from Ingot 1 are shown in Figure III-6 and Figure III-7, taken at five inches and seven inches from the chill, respectively. In these figures a section parallel to heat flow is shown, matched at a edge to the section parallel to the chill surface. Cooling curves from Ingot 1 are shown in Figure III-8.

Dendrite Growth Forms

Figure III-9 shows another section of Ingot 1 at five inches from the chill, in which photomicrographs of the longitudinal and transverse structures have been mounted on a block of wood. A photograph of the block shows the three-dimensional aspect of the dendrite structure. In the transverse section, the light etching regions (which are low solute, first solidified regions) have a cross type appearance. The more prominant crosses are traces of the primary dendrite growth forms. In the longitudinal section, other traces are visible. These traces can be matched with traces in the vertical section. In fact, the primary dendrite growth form is sheet-like, so that the linear dendrite traces in the longitudinal and transverse sections represent the intersection of dendrite sheets with these two sections.²⁹

Secondary dendrite growth forms show up as approximately horizontal lines in the longitudinal section. They are also sheet-like but
Figure III-5: Transverse sections from water chilled Columnar Ingot 1. (a) 1/2 inch from chill (b) 1 inch (c) 1-1/2 inches (d) 2 inches (e) 2-1/2 inches (f) 3 inches (g) 3-1/2 inches (h) 4 inches (i) 4-1/2 inches (j) 5 inches (magnification 12X).
Figure III-5 (continued): (k) 5-1/2 inches (l) 6 inches (m) 6-1/2 inches (n) 7 inches (o) 7-1/2 inches (p) 8 inches (q) 8-1/2 inches (r) 9 inches.
Figure III-6: Longitudinal (top) and transverse (bottom) sections of Columnar Ingot 1 at 5 inches from the chill. Note that some primary sheets have a "line" type structure while others show a "cross" type structure (composite photograph, magnification 12X).
Figure III-7: Transverse (bottom) and longitudinal (top) sections of Columnar Ingot 1 at 7 inches from the chill. Note in the transverse section that tertiary sheets are often separated from primary sheets by a high solute region. The connection between secondary and tertiary sheets is evident in the transverse section (composite photograph, magnification 12X).
Figure III-1: Cooling curves for water with columnar ingots.
Figure III-9: Columnar dendrite structure at five inches from the chill in Columnar Ingot 1. Composite three-dimensional photograph of transverse and longitudinal sections mounted on a block of wood (approximate overall magnification 30X).
imperfect. A transverse section often cuts through a secondary sheet, revealing broad light etching areas of low solute concentrations. Tertiary growth forms might also be characterized as sheet-like, although their thickness is a significant fraction of their width, and neither "width" nor "thickness" is constant. Tertiary sheets are most often separated from primary sheets by a high solute region, indicating that they grew from the secondary sheets and not from the primary sheets. Refer to Figure I-1 for the orientations of the primary, and tertiary sheets. A schematic etched structure is given in Appendix H.

**Dendrite Sheet Orientation**

Dendrite sheets are oriented in $\{100\}$ planes of the dendritic crystal. In columnar growth the dendrites tend to become aligned with a $\langle 100 \rangle$ direction in the heat flow direction. The $\langle 100 \rangle$ direction is the intersection of two primary dendrite sheets. It is possible to calculate the angle between the dendrite direction $\langle 100 \rangle$ and the vertical direction from the orientation of dendrite traces in two perpendicular sections, like those in Figure III-9. Vector analysis is used.

The dendrite trace is the intersection of a dendrite sheet with the plane of polish. The appearance of the trace depends in two ways on the angle between the dendrite sheet and the plane of cut. First, in a longitudinal section the angle $\gamma$ between the dendrite trace and the vertical direction can vary depending on the cut. When the plane of cut is at a low angle to the dendrite plane, $\gamma$ will be near 90°. When the plane of cut is near 90°, $\gamma$ will be equal to the angle $\theta$
between the heat flow direction and the dendrite sheet. Second, the dendrite trace width depends on the angle between the dendrite sheet and the plane of cut. When the plane of cut is near 90°, the trace width equals the sheet thickness. At low angles of cut, the trace width is much greater than the sheet thickness. See Appendix G for an analysis of (1) trace angle \( \gamma \) versus sheet orientation \( \theta \) and cutting angle and (2) trace thickness versus angle of cut. The traces in the longitudinal section of Figure III-9 are formed by the plane of cut intersecting the dendrite sheets at about 45°. The trace thickness is somewhat greater than the trace width visible in the transverse section, as expected. In Figure III-4 several very wide traces of "bands" result when the dendrite plane lies almost in the cutting plane (labelled "4").

**Dendrite Elements**

In Figure III-9 several dendrite crosses of essentially identical orientation exist within a single grain. The large sections of Figure III-6 and Figure III-7 show this more clearly. All of the crosses of a single orientation are part of the same dendrite or grain. However, it is clear from the picture that the primary dendrite sheets are broken between the dendrite crosses. These separated regions of a dendrite are called dendrite elements. A schematic picture of dendrite elements is shown in Figure II-1a.

Pictures of chill structures like those in Chapter II show that there is a continuous path of low solute near the chill region. In other words, there is a path to get between any two points in the structure without crossing a sub-boundary. In the case of a water chill ingot, this
connection in a transverse section disappears in most grains very near the chill (see Figure III-5) as dendrite crosses grow up into the ingot. In this case, each dendrite cross is an independent dendrite element. Several dendrite crosses may exist in a dendrite element, as discussed later for air cooled ingots. Sometimes a single dendrite sheet, rather than a cross, forms the basis of a dendrite element. This produces a "line" type structure (See Figure III-5q). One striking piece of evidence showing the independence of dendrite elements is their intermingling between grains. A dendrite element or group of dendrite elements from one grain may be found in the midst of elements of a neighboring grain.\textsuperscript{3,49} In a macrosection, intermingling causes very jagged and broken grain boundaries (see Figure III-2) rather than the smooth boundaries which are typical of a pure metal.

b. Air Chilled Ingot

Transverse sections from Ingot 2 are shown in Figure III-10. The cooling curves from Ingot 2 are given in Figure III-11.

Dendrite crosses are not so obvious in Figure III-10 as they are in Figure III-5. The structures show that the primary growth forms are very coarse and not so regular as in the water chilled ingots. The microstructures are not only coarser but also more disconnected. Perhaps dendrite elements associated with the primary growth forms could themselves be described as an array of dendrite elements on a finer scale.
Figure III-10: Transverse sections from air chilled Columnar Ingot 2. (a) At chill (b) 1/2 inch (c) 1 inch (d) 1-1/2 inches (e) 2 inches (f) 2-1/2 inches (g) 3 inches (h) 3-1/2 inches (i) 4 inches (j) 4-1/2 inches (magnification 12X).
Figure III-10 (continued): (k) 5 inches (l) 5 1/2 inches (m) 6 inches (n) 6 1/2 inches (o) 7 inches (p) 7 1/2 inches (q) 8 inches (r) 8 1/2 inches (s) 9 inches.
Figure III-11: Cooling curves from air chilled Columnar Ingot 2. The ingot was water quenched after solidification was complete.
Sections from Ingot 3 and associated cooling curves have been described previously. The cooling rates in this air cooled ingot were greater than those in Ingot 2. The primary growth form structure and dendrite elements differ somewhat from the water chilled case. Near the chill extensive interconnection exists between dendrite crosses. The connection in a transverse section gradually breaks down until at 3 inches from the chill each primary growth form cross is part of a separate dendrite element.

4. Dendrite Growth Form Spacing

Primary spacing in a columnar ingot is measured in a transverse section. It is the average distance between primary sheets, the direction of measurement lying perpendicular to the dendrite trace spacing measured. Since the primary growth forms are nearly perpendicular to the polished surface, the distance between the dendrite traces will be an accurate measure of primary dendrite sheet spacing. In the case of a cross-type structure both perpendicular directions are measured and an average taken. In the case of a "line" structure, and spacing is taken perpendicular to the "lines".

Secondary spacing is measured on a longitudinal section. Often a duplex secondary spacing exists. Near the primary sheets, the secondary sheets have one spacing. Further from the primary sheets, and higher in the ingot, the secondary spacing is often greater by a factor of 2 or 3 because some secondary sheets stop short in their growth. On the other hand, in some locations the finer spacing is maintained. See Figures III-6 and III-7.
Tertiary spacing is measured in transverse sections, like those of Figure III-8. The tertiary spacing is the characteristic spacing of the structure, because it represents the spacing which is most typical of the distance between low solute areas (or twice the distance between high and low solute areas). See Appendix H for a schematic diagram of dendrite growth form spacing measurements.

Primary, secondary and tertiary dendrite sheet spacing from Columnar Ingot 1 is plotted versus distance from the chill in Figure III-12. A best curve has been drawn through the data for each order of spacing. Cooling curves at various distances from the chill for Columnar Ingot 1 are given in Figure III-8. In Figure III-13 spacings of dendrite growth forms in water chilled Ingot 1 obtained from the best curves through the data of Figure III-12 have been plotted against solidification time. For the purposes of this thesis solidification time is defined as the time for cooling from the liquidus temperature to 50°F below the liquidus temperature. Table J-1 gives solidification times for Ingots 1, 2, and 3.

Spacings from air cooled Ingot 3 (see Table J-2) were plotted in Figure III-13. Primary growth form spacings are plotted as solid squares. Secondary and Tertiary spacings are plotted together as open squares. In the case of air cooled Ingot 2, primary growth form spacings were variable between about 1000 to 2000 microns (see Figure III-10). These are not plotted; if plotted, they would scatter about the horizontal dashed line of Figure III-13 from 2000 to 5000 seconds. There was little change in cooling rates in Ingot 2 from chill to top; solidification time at the chill in this Ingot was longer than that at 10 inches from the chill in
Figure III-12: Dendrite growth form (sheet) spacing versus distance from the chill in water chilled Columnar Ingot 1. Symbols near primary spacing data points indicate cross versus line type structure.
Figure III-13: Dendrite sheet spacing versus solidification time for dendrites grown under various conditions. Solid symbols are primary, open symbols are secondary or tertiary. Solidification time was taken as the time for cooling from the liquidus to 50°F below the liquidus.
air chilled Ingot 3. Tertiary spacing varies little in the ingot, from about 200 microns at the chill to near 300 microns at 9 inches from the chill (see Figure III-10). Tertiary spacings are plotted in Figure III-13 for the distances from the chill where thermal data was taken.

Primary and tertiary spacing from an equiaxed ingot described in Chapter VI (see Figure VI-3) are plotted in Figure III-13 as triangles.

Primary growth form spacing from seeded dendrites grown in a single crystal furnace (discussed further in Chapter IV) have also been plotted in Figure III-13. Solidification occurs at a constant rate of temperature change in these crystals. Cooling conditions, therefore, differ from unidirectional solidification. A different trend develops in primary growth form spacing; the spacings are markedly lower at longer solidification times, and primary growth form spacing depends little on solidification time. Temperature gradients were almost constant during freezing of these crystals (see Table IV-1).

Best curves drawn through the data of Figure III-13 give the following results. In water cooled Columnar Ingot 1 primary growth form spacing is proportional to the square root of freezing time, in agreement with previously reported data on "dendrite arm spacing". For secondary and tertiary growth form spacings in all the ingot considered, spacing is proportional to a lower power of solidification time than primary spacing. The line shown in Figure III-13 has a slope of .28. Higher order spacing is thus proportional to the fourth root of solidification time (in agreement with the results of Norweth and Mendum).
5. Interrupted Solidification of the Air Cooled Ingot.

One ingot was made by air cooling until the liquidus isotherm was about 3/4 inch from the chill. At this time water was turned on and the structure was effectively quenched in. Cooling curves for this ingot are given in Figure III-15. Figure III-14 shows transverse structures at 1/2 inch from the chill, 5/8 inch from the chill and 3/4 inch from the chill. The coarse dendrite traces are the regions which were solid before water quenching. The other areas have a dendrite spacing typical of water cooling. (Segregation studies made on this ingot are discussed in Chapter VII.)

D. Discussion

1. Development of Preferred Orientation.

In the introduction it was shown that dendrites in columnar growth tend to develop a $\langle 100 \rangle$ preferred orientation. The structure of columnar dendrites help to explain this orientation. First $\langle 100 \rangle$ dendrite sheets tend to form in columnar grains and are particularly well developed perpendicular to the chill. Second, high solute regions (high solute liquid during growth) separate dendrite elements within a dendrite. Both of these factors contribute to anisotropic thermal conductivity in a columnar dendrite. Solid has a better conductivity than liquid; increase in solute lowers the conductivity.

For this reason alone, once a dendrite forms with sheets in any direction, conductivity will be highest in that direction. In addition, the formation of independent dendrite elements reinforces the anisotropy
Figure III-14: Photomicrographs of air-water cooled Columnar Ingot 4. (a) 1/2 inch from the chill, transverse section, (b) 5/8 inch from the chill, (c) 3/4 inch from the chill. (Magnification 12X)
Figure III-15: Cooling curves from Columnar Ingot 4, air cooled and water quenched. The temperature inversion at the chill is due to solidification shrinkage; the casting shrinks from the chill surface, reducing heat transfer efficiency.
of conductivity. Dendrite sheets may be extensively developed in one 
\(\langle 100 \rangle\) direction which is nearly perpendicular to the chill but 
separated by high solute regions in the two other \(\langle 100 \rangle\) directions.

Compare two grains, one oriented perpendicular to the chill, the 
other having a \(\langle 100 \rangle\) direction at an angle to the chill. Even if 
dendrite sheets are well developed in the second grain, heat will have 
to travel further along them.\(^{29}\) (Except for one \(\langle 100 \rangle\) sheet which is 
perpendicular to the chill for grains on the \(\langle 100 \rangle\) zone). The better 
oriented grain should grow ahead of the more poorly oriented one 
because of the difference in conductivity.\(^{29}\)

2. Formation of Dendrite Sheets.

It is also necessary to explain why dendrites tend to develop a 
sheet-like morphology. Flemings has suggested that the \(\langle 100 \rangle\) planes 
may be slow growing planes.\(^{50}\) Chalmers has suggested that heat flow 
in columnar grains tends to favor a sheet-like structure.\(^{50}\) He feels 
that the gradients set up in a rod-like dendrite structure cause a 
more rapid growth at the junction of primary and secondary dendrite 
rods which may then "fill in". However, sheet-like secondary growth 
forms are observed which lie parallel to the chill surface, 90° to 
macroscopic heat flow. Also sheet-like growth forms are produced under 
very low gradients in equiaxed grains (see Figure VI-3). Flemings\(^{3}\) 
has suggested another explanation for sheet formation based on surface 
energy considerations. Surface energy is high in a rod-like morphology 
and filling in will occur at the corners in preference to other areas.

Figure III-16 shows two $[100]$ dendrite axes growing into the liquid. It shows the dendrite sheet as a web growing in the plane of the paper. Two other dendrite planes intersect this sheet perpendicular to the plane of the paper. The axes formed by the intersections of the perpendicular sheets extend into the liquid. There are two possible types of growth into the liquid. Consider the web joining the two dendrite axes. If the dendrite sheet is to remain free of sub-boundaries, the base of the web must grow forward rapidly enough to "zip up" the dendrite sheet. The base cannot be left behind. That is, if the sides grow together (shown as velocity $V_2$ in Figure III-16) before the base grows up, (velocity $V_3$), then a sub-boundary will form. The two sides growing toward each other will not join each other. In fact, solute rejection from each will cause them to "avoid" each other. Other factors must be considered to predict cases in which the base will grow up and close the web and cases in which this will not occur. Secondary growth forms on the primary sheets will complicate the picture but it is not clear what effect this might have.

Whatever the cause, if the base is unable to zip up, disconnection occurs; a dendrite element results from disconnections on all four sides of a single primary dendrite axis, which is the basis of the dendrite cross. No misorientation need be caused by the formation of independent elements, per se. Once independent elements form, however, their misorientation may be caused by several factors. They are long thin independent projections into the liquid. Differential contraction or
Figure III-16: Schematic sketch of a growing dendrite showing a mechanism for the formation of dendrite elements. For the sake of clarity, only the primary dendrite sheets are shown. One sheet is in the plane of the paper; orthogonal sheets are represented by the vertical lines. The intersection of the two sheets is the <100> dendrite direction, which projects into the liquid. The dendrite sheet in the plane of the paper may solidify in two ways, as shown above. In one case, the sheet grows quickly at its root and remains continuous (V₃ is large). In the other case, the sides grow together before the root is able to grow past (V₂ is large). In this case, solute rejection from the two sides causes them to avoid each other and a boundary or separation forms in the sheet. This is the origin of the dendrite element boundaries.
convection in the liquid might cause enough deformation to produce the observed misorientation.


The results on dendrite growth form spacing show that primary dendrite growth form spacing has a different dependence on solidification time from higher order growth form spacing (secondary and tertiary). Under some conditions, primary growth form spacing is proportional to the square root of solidification time. Higher order growth form spacing, on the other hand, is more nearly proportional to the fourth root of solidification time. The differing dependence on solidification time may be a reflection of the mechanisms involved in the determination of primary and higher order spacings, which will now be discussed.

First, assume that primary growth forms have developed, and consider secondary branching. Secondary branches will form consecutively on the primary growth form as it grows forward into the liquid. The newest "branches" to grow out will be the furthest from the chill. The spacing of these branches will depend in some undetermined way on constitutional supercooling and interface energy. The supercooling conditions depend on both cooling rate and the morphology of existing solid. For example, the existence of one branch will influence the environment of the region where the next branch would be expected. Each branch will develop according to the local conditions which exist when it forms. Thus, the spacing which results can adjust almost continuously to changing growth conditions at various heights in the ingot.
There can be no such continuous change in primary growth form spacing. Primary growth forms are units whose spacing has already been established before solidification in any region. Their growth into a region occurs not by branching, but by growth onto already existing solid. Primary growth form spacing does change, however, under some conditions. There are two ways in which an increase in spacing may be achieved, as observed in water cooled Columnar Ingot 1.

1. The elimination of individual primary sheets. This is possible, but not sufficient to account for the entire change in spacing.

2. The elimination of whole dendrite elements. Sheets or elements may become unstable and stop growing. Thus, change in primary spacing is a discontinuous process. (Hultgren\textsuperscript{12} suggested a more continuous process with alternate dendrite elements being suppressed). If a primary growth form is eliminated, then a "vacancy" is left, and the morphology of primary growth forms in neighboring dendrite elements will adjust to fill the gap. This type of mechanism helps to explain the non-uniform spacing of primary dendrite growth forms. This is contrasted to the regular spacing observed in higher order growth forms (at the place of branching).

The mechanism of primary growth form spacing must now be considered. Independent projections advance into the liquid. The presence of one growth form affects the others through the liquid, by heat and solute diffusion. (Since the effects of heat diffusion are comparable to solute diffusion, and because heat diffusion should be so rapid over these distances that no gradients exist, only solute diffusion is considered here.) At greater distances from the chill, where the
solidification time is longer, there is more time for diffusion of solute from an advancing dendrite element tip. Therefore, there is a greater influence of one dendrite element on the environment of its neighbor (see Figure III-17). One dendrite element might fall behind its neighbors either randomly or for some definite reason (because it is a poorer heat conductor, for example). At high cooling rates it would be undercooled and grow forward before side diffusion from its neighbors had influenced its progress. But at a longer solidification time (high in the ingot), solute rejected by its neighbors would tend to diffuse sideways and pile up ahead of the backward element, further suppressing its growth. Thus a dendrite element which was behind its neighbors for whatever reason, would become unstable and cease to grow. This is called here "terminal instability". Terminal instability occurs between chill surface structures which do not contain dendrite crosses (primary growth forms) and structures very close to the chill surface (compare Figure III-8 with Figure III-5, see also Figure III-3). Terminal instability also may occur in secondary sheets (some of these are quickly eliminated near the primary sheets; see Figures III-6 and III-7). Terminal instability is not "choking off" in the usual sense; "choking off" involves the physical barrier presented by one growth form which lies in the path of growth of another growth form.
Figure III-17: Mechanism of change in primary dendrite growth form spacing during unidirectional solidification (schematic). (a) A group of dendrite axes projecting into the liquid. Arrows indicate solute rejection. (b) One axis falls slightly behind the others. Solute rejected by its neighbors impedes its growth. (c) Suppression of the projection is complete as side branches form on neighboring axes. Note: The axes are tips or dendrite elements within a single grain.
The Effect of Gradients

It is of interest in connection particularly with the results of Figure III-13 for single crystal dendrites, to consider the effect of gradients on terminal instability. For longer solidification times, single crystal dendrites had a smaller spacing than the columnar dendrites. Also, the spacing was not dependent on solidification time. The thermal gradients during growth of these crystals were about constant. Possibly the relatively high thermal gradients (relative to the cooling rate) prevent the primary growth forms from "getting out of line". Dendrite elements which fall incrementally behind become undercooled and grow faster. Thus side diffusion is less effective and terminal instability is prevented. With a smaller primary growth form spacing (higher side surface area), the primary growth forms are less unstable with respect to branching. Thus, they are thicker and have fewer branches (see Chapter IV). The cellular structure has this feature, so terminal instability may be the key to the cellular-dendrite transition.

In summary, the mechanism of primary spacing must result from an interplay between the stabilizing effect on many dendrite growth forms of heat removal, balanced against growth form interaction by solute diffusion. On the other hand, higher order spacing is a consecutive effect, in which one growth form spacing is determined at a time. Primary spacing adjusts discontinuously to a changing cooling rate, while secondary spacing can change continuously.
E. Summary

Columnar dendrite structures produced under various conditions have been examined. The dendrite growth forms have been shown to be largely sheet-like (see Figure III-9). Dendrite axes are seen in a longitudinal section, while dendrite "crosses" or "lines" are typical of transverse section. The difference between primary growth form spacing and secondary growth form spacing has been discussed. The primary growth form spacing dependence on solidification time differs from that of the secondary growth form spacing, reflecting a different mechanism of instability.

Secondary growth forms are produced consecutively during solidification and their spacing can adjust to the changing thermal conditions. Primary growth form spacing, on the other hand, must increase discontinuously with the elimination of growth forms during growth from the chill (see Figure III-17).

Four structural zones may exist near a chill surface in castings which have no distinct "chill zone" (see Figure III-3). The first is a high solute exuded layer formed late in solidification.

The second zone is a low solute layer at the chill surface caused by the growth of two types of dendritic growth forms. One type is projections onto the chill surface of \( \langle 100 \rangle \) directions of the dendrite. These exist as "rods" at the chill surface. The other type of dendrite growth form at the chill surface is an intersection of a \( \langle 100 \rangle \) dendrite sheet, usually a sheet lying at high angle to the chill surface. These sheets grow both along the chill surface and into the casting from the chill surface.
The third zone near the chill is formed by \( \gtrless 100^\circ \) dendrite sheets which are at a low angle to the chill surface. These sheets do not intersect the chill surface, unlike the dendrite sheets in the second zone. In zones two and three substantial growth takes place transverse to the heat flow direction.

Zone four is the typical columnar structure which results from the extensive growth of orthogonal dendrite sheets, both of which lie at a relatively high angle to the chill surface. All four "zones" in the chill region may exist in a single dendrite.

Independent dendrite "elements" form during growth away from the chill (Figures III-6, III-7, and III-9). A mechanism of formation of dendrite elements, has been proposed (see Figure III-16). Under conditions of unidirectional air cooling, dendrite elements may consist of more than one dendrite "cross". Normally, however, each dendrite cross is a separate dendrite element.

Misorientation develops between dendrite elements during growth from the chill. Several reasons for the misorientation have been discussed. Segregation between dendrite elements (within a dendrite) produces high solute boundaries very similar to grain boundaries. In some cases the amount of second phase at dendrite element boundaries is equivalent to the amount of second phase at grain boundaries (see Figures III-6 and III-7).

An explanation of the development of preferred orientation based on anisotropic heat flow in dendrite plates and dendrite elements has been discussed.
IV. SEEDED DENDRITE EXPERIMENTS

A. Introduction

There are several features of the growth of columnar dendrites which are not fully revealed in the unidirectional solidification experiments discussed in the previous chapter. First, the dendritic structure has been studied in detail only for dendrites grown with \( \langle 100 \rangle \) directions nearly in the heat flow direction. Dendrites in other orientations are usually suppressed by their more favorable oriented neighbors before they reach a size suitable for study. Second, the spacing of dendrite growth forms and the order of branching has not been studied under controlled conditions which allow a separation of critical parameters. Finally, the mechanism of development of preferred orientation in columnar growth studied to date only by Walton and Chalmers is not completely understood.

The first problem mentioned above, namely the determination of the structure of dendrites far from a \( \langle 100 \rangle \) orientation, is the most easily solved. It should be emphasized that there is a difference between the structure of a dendrite that has grown in a \( \langle 100 \rangle \) direction sectioned on its \( \{110\} \) or \( \{111\} \) planes and the structure of a dendrite that has grown in a \( \langle 110 \rangle \) or a \( \langle 111 \rangle \) direction sectioned on a (110) or a (111) plane. The former has been studied to some extent; the latter has not. If a seed of known orientation is used, and growth caused to proceed from this seed, a single crystal of the same orientation should be obtained. If growth conditions are favorable for dendritic growth, a dendritic crystal of the desired orientation should be obtained. No other crystals
should be present. In practice, however, stray crystals are often nucleated and may suppress growth of the seeded crystal.

The second problem is more complex. The solution involves a study of the parameters controlling dendrite element spacing and the development of primary, secondary, and higher order dendrite growth forms. First the important parameters must be isolated. These may include thermal gradients in the solid and liquid, diffusion in solid and liquid, liquid-solid surface characteristics, cooling rate over various parts of the solidification range, solute content and phase diagram characteristics. Orientation of the dendrite may also influence dendrite growth form spacing and order of dendrite element branching.

The last problem considered here is the development of preferred orientation. For a preferred orientation to develop in columnar grains, grains of one orientation or range of orientations must suppress all other grains. In aluminum and other face centered cubic metals, as mentioned previously, a \(\langle 100 \rangle\) preferred orientation develops. It is of interest to determine how a grain of one orientation suppresses a grain of another orientation. Walton and Chalmers\(^{23}\) have performed several bicrystal and one tricrystal experiments which are pertinent to this discussion. In the tricrystal experiment they placed a \(\langle 100 \rangle\) seed in the middle and two \(\langle 111 \rangle\) seeds on either side in an especially designed boat. The metal was 99.999 per cent lead. After melting back to the three seeds the furnace was pulled away. The \(\langle 100 \rangle\) dendrites suppressed the \(\langle 111 \rangle\) dendrite from both sides. No dendritic structure was observed, however, due to the low impurity level. In two bicrystal experiments a seed with
a $\langle 100 \rangle$ direction in the heat flow direction was placed on either side of a seed with a $\langle 100 \rangle$ direction 30° from the heat flow direction. (A complete specification of the orientation of the two seeds was not given. The $\langle 100 \rangle$ seed, for example, might be aligned with a $\langle 100 \rangle_2$ or a $\langle 110 \rangle_3$ plane parallel to the other seed.) In these experiments the lead was alloyed with .02 per cent silver. The growth rate was .01 centimeter/second and the gradient estimated as 20°C/centimeter. Walton and Chalmers found that the $\langle 100 \rangle$ dendrite suppressed the second dendrite when their $\langle 100 \rangle$ directions were diverging, but that grain boundary deviation was less marked for the converging dendrite systems. Walton explained that the grain boundary deviation in the tricrystal was due to thermal supercooling, while in the bicrystal experiments dendrites were produced by constitutional supercooling. In the former case, "The mechanism by which they are eliminated depends on the production of latent heat by the more favorable oriented dendrites. This warms the liquid behind them thereby interfering with the growth of less favorably oriented dendrites." Fifty "The mechanism by which unfavorably oriented grains are eliminated . . . in the latter case . . . depends upon the production of new dendrites . . . [dendrite elements] . . . at a grain boundary between two diverging dendrite systems." Walton thus points out the essential difference between converging and diverging systems: in diverging systems, new dendrite elements must be produced, while in converging systems existing dendrite elements must be eliminated.
In two other bicrystal experiments, Walton investigated the effect of growth conditions on the development of preferred orientation. A lead +0.01 per cent silver bicrystal was grown with diverging dendrite growth forms as in the experiment discussed previously. In this case, however, the growth rate was reduced to 0.001 centimeter/second and the gradient increased to an estimated 100°C/centimeter. Less grain boundary deviation was noted under these conditions than in the previous experiment. The second bicrystal was lead containing 5 x 10^-3 per cent silver. "The first half of the specimen was grown very slowly with the high temperature gradient. Then the air was shut off and the rate of growth increased. The result was that the first half was grown with a cellular interface which changed to dendritic growth in the last half." A sharp change in grain boundary direction occurred between the first and last halves attributed, by Walton, to dendrite branching.

B. Apparatus and Procedure

1. Castings.

Aluminum-4.5 per cent copper alloy or 99.99 per cent aluminum was melted in an electric furnace, heated to 1500°F and poured into graphite molds. These molds were coated with amorphous carbon and preheated to 300°F to increase fluidity. Castings molds (shown in Figure IV-1) were of two designs. In one the cross section was 1/4 inch by 1/4 inch along the entire length. In the other the section increased to 1/4 inch by 1/2 inch from 1/4 inch by 1/4 inch. Thus the polycrystalline casting blanks cast into these molds also had these dimensions.
Figure IV-1: Casting mold designs. Ruler indicates magnification. Seed mold (top), alloy mold (center), cover (bottom).

Figure IV-2: Growth mold designs. Seed mold (top), alloy mold (center), cover (bottom).

Two growth molds are shown in Figure IV-2. In order to obtain an alloy single crystal a seed was put in one end of the mold. A polycrystalline casting was fitted into the remainder of the mold and a graphite cover tied on so as to expose the seed and the seed-casting junction. The casting was melted in a boat-type single crystal grown furnace of Chalmer's design and melting allowed to proceed back to the seed. An argon atmosphere was used. After the seed had started to melt, the oxide layer between the seed and the casting-blank was broken with a Fiberfrax coated steel wire and the furnace moved so as to allow freezing to start. After freezing had proceeded under the cover, the furnace was tilted so that growth proceeded up a 30° incline.

In most cases, a water cooled copper chill was placed in contact with the graphite at the seed end of the mold. Growth rates were varied by means of a variable speed motor which pulled the furnace past the specimen. Temperature gradients were measured by means of chromel alumel thermocouples whose protection tubes passed to intervals through the boat cover into the melt.


Pure aluminum (99.99 per cent) crystals of a desired orientation were made in much the same way as alloy single crystals. When a seed of a new orientation was required, a single crystal was grown from another seed in one of several graphite boats designed to change and correct the crystal orientation. In these boats the axis of the seed was at an angle
of from 0° to 45° from the axis of the crystal being made. (For an
angle of 0°, the orientation change was a rotation about the axis.) Two
inch seeds were cut from the new crystal. The specimens were cut with a
jeweler's saw and the deformed metal removed by etching in concentrated
Keller's etch.

4. Orientation.

The designation \( \langle 100 \rangle \{110\} \) indicates a crystal having a \( \langle 100 \rangle \)
direction parallel to its long axis and a \( \{110\} \) plane parallel to its
horizontal surface. This designation is used throughout for both seeds
and alloyed single crystals. (A designation may be given such as
\( \langle 110 \rangle \{110\} \{100\} \); in this case the final \( \{100\} \) indicates that a
\( \{100\} \) plane is parallel to the vertical side surface. This information
is sometimes convenient, but also redundant since the crystal orientation
is completely determined without it.) Orientations were determined by
the Laue back reflection method.

C. Experiments and Results

Seed crystals were made with \( \langle 100 \rangle \), \( \langle 110 \rangle \), and \( \langle 111 \rangle \) directions
along their long dimension. Dendritic grains of aluminum-4.5 per cent
copper were grown from these seeds. A list of orientations studied, the
furnace speed, average cooling rate, solidification time, primary
dendrite growth form spacing and temperature gradients are given in
Table IV-1.
TABLE IV-1
Single Crystal Dendrite Data

<table>
<thead>
<tr>
<th>Dendrite Orientation</th>
<th>Speed of furnace (cm/hr)</th>
<th>Gradient in Liquid (°C/cm)</th>
<th>Cooling Rate (°C/sec)</th>
<th>Solidification Time (sec)</th>
<th>Average Primary Dendrite Spacing***</th>
</tr>
</thead>
<tbody>
<tr>
<td>(100) (100)</td>
<td>9.4</td>
<td>21</td>
<td>.062</td>
<td>450</td>
<td>365</td>
</tr>
<tr>
<td>(100) (110)</td>
<td>13.3</td>
<td>23</td>
<td>.096</td>
<td>290</td>
<td>345</td>
</tr>
<tr>
<td>(110) (100) (110)</td>
<td>9.4</td>
<td>17</td>
<td>.060</td>
<td>465</td>
<td>385 &quot;lines&quot;</td>
</tr>
<tr>
<td>(110) (110) (100)</td>
<td>9.2</td>
<td>14</td>
<td>.056</td>
<td>500</td>
<td>475 &quot;lines&quot;</td>
</tr>
<tr>
<td>(110) (100) (110)</td>
<td>76.7</td>
<td>18</td>
<td>.39</td>
<td>710</td>
<td>320 &quot;lines&quot;</td>
</tr>
<tr>
<td>(111) (110) (112)</td>
<td>9.4</td>
<td>19</td>
<td>.62</td>
<td>450</td>
<td>Not measured</td>
</tr>
<tr>
<td>(100) (110)</td>
<td>3.1</td>
<td>19</td>
<td>.023</td>
<td>1200</td>
<td>263</td>
</tr>
</tbody>
</table>

* Structure not shown in thesis.

** Defined in Chapter III.

*** Several dendrites had different spacings in the vertical and horizontal directions. An average value is given here, but both values are plotted in Figure III-13. "Line"-type structure spacings were determined perpendicular to the lines. (Microns)

The specimens were sectioned, polished and etched. Sections perpendicular to the long axis of the specimen were approximately perpendicular to the heat flow direction during growth, while those along the specimen surface were parallel to the heat flow direction. Some transverse component of the temperature gradient from the top (the boat cover) to the bottom (the boat) probably existed, however, although this was not measured; the boat was in direct contact with the water chill, while the cover was not, so the boat would tend to be colder than the cover at any point.
1. \textbf{\langle 100 \rangle Dendrites}

Figure IV-3 shows a \textbf{\langle 100 \rangle} dendrite grown from a \textbf{\langle 100 \rangle \{100\}} seed. Several features are of interest. The primary dendrite sheets are extended vertically more than horizontally, and the eutectic segregation pattern is more continuous vertically than horizontally. This may be caused by the component of the temperature gradient in the vertical direction. Also, the primary dendrites are quite thick and secondary dendrite growth forms are not well developed. In fact, a section polished parallel to the bottom surface (Figure IV-4) shows no evidence of dendrite sheets oriented perpendicular to the heat flow direction. Near the top the structure consists of close packed dendrite elements having only primary growth forms, while toward the bottom there are secondary dendrite growth forms between the thicker primary sheets. The secondary dendrite growth forms, like the primary, seem to be sheets oriented in the heat flow direction. The minimum dendrite spacing changes little from top to bottom in the same section. Although this is essentially a single crystal, dendrite misorientation of up to 10° can be seen in adjacent regions. The origin of this misorientation may be in the seed ("lineage" or "striations") or a result of distortion during growth. A stray grain can be seen just to the right of center in Figure IV-3.

Figure IV-5 shows a \textbf{\langle 100 \rangle} dendrite grown from a \textbf{\langle 100 \rangle \{110\}} seed. In this case the primary dendrite sheets were approximately equally well developed in both directions, and there is less misorientation between dendrite elements than in the \textbf{\langle 100 \rangle \{100\}} dendrite. Again, however, the top of the structure is close packed primary dendrite growth forms,
Figure IV-3: \( \{100\} \bigcap \{100\} \) dendrite (magnification 12X).

Figure IV-4: Section parallel to the bottom surface of the dendrite in Figure IV-3 (magnification 12X).
Figure IV-5: $\langle 100 \rangle \begin{Bmatrix} 110 \end{Bmatrix}$ dendrite (magnification 12X).

Figure IV-6: $\langle 110 \rangle \begin{Bmatrix} 100 \end{Bmatrix} \begin{Bmatrix} 110 \end{Bmatrix}$ dendrite (magnification 12X).
while secondary growth forms are more evident toward the bottom; minimum
dendrite spacing is similar from top to bottom. Although an odd stray
grain may be found, none developed significantly in either the $\langle 100 \rangle$
$\{110\}$ crystal or the $\langle 100 \rangle \{100\}$ grain discussed previously.

2. $\langle 110 \rangle$ Dendrites

Figure IV-6 shows a $\langle 110 \rangle \langle 100 \rangle \{110\}$ dendrite near the junction
with the seed. Note the stray grains in the upper part of the structure.
This upper structure is produced during nabbling to break the oxide film
and is a bulge on the specimen. The $\langle 110 \rangle$ dendrite is in the lower
part of the picture. It consists of well-developed dendrite "lines"
horizontally and other less well-developed forms vertically. The
horizontal "lines" are $\langle 100 \rangle \{110\}$ dendrite planes oriented perpendicular to
the growth direction. Figure IV-7 is a section taken 3-1/4 inches from
the seed. Several stray grains can be seen in the upper part of this
section (at 6 inches from the seed the stray crystals account for 60 per
cent of the cross section). The horizontal dendrite sheets throughout
this crystal, although pronounced, are somewhat broken (as in Figure IV-7).

To minimize the possible effect of transverse (vertical) thermal gradients
a $\langle 110 \rangle \langle 110 \rangle \{100\}$ dendrite was grown. In this case the $\langle 100 \rangle \{100\}$
dendrite planes which lie perpendicular to growth are vertical. Even
with a vertical component to the temperature gradient, the gradient will
still lie in the $\langle 100 \rangle \{110\}$ planes. Figure IV-8 shows the structure of the
dendrite near the seed junction. The structure in the bulge (at the top)
illustrates the difference between the dendrite structure of a dendrite
grown in a $\langle 110 \rangle$ direction sectioned on a $\langle 110 \rangle \{110\}$ plane (the lower part
of the picture) and the dendrite structure of a dendrite that has grown
in a $\langle 100 \rangle$ direction sectioned on a $\langle 110 \rangle \{110\}$ plane (the upper part
Figure IV-7: Dendrite seeded $<110> \approx 100^\circ$, 3-1/4 inches from seed (magnification 12X).

Figure IV-8: $<110> <110> \approx 100^\circ$ dendrite (magnification 12X).
of the picture). The growth direction in the upper part was more nearly in a (100) direction because growth was upward into the bulge. In this case two sets of (100) planes developed and both can be seen in the section. The plane of the horizontal dendrite traces lie at 45° to the plane of cut and are apparently thicker for this reason than are the vertical dendrites (see Figure G-2 and H-1).

The structure of the upper part of the figure is more broken than that in the lower part. This is additional support for the idea that intersecting dendrite plates result in the formation of independent dendrite elements (see Figure III-16). Figure IV-9 shows a section of the same crystal at 1-1/2 inches from the seed. A stray crystal can be seen at the left. (At 5-1/2 inches from the seed two strays occupied about 25 per cent of the cross sectional area.) Notice how many of the vertical dendrite sheets are nearly perfect, unbroken from the top to the bottom of the specimen, in contrast to the horizontal dendrite sheet structure of Figure IV-7. Flemings has suggested that natural convection currents may be responsible for the difference in structure between the (110) (100) dendrite and the (110) (110) dendrite. A vertical component of the temperature gradient might also account for the difference, as mentioned previously.

Figure IV-10 shows the structure of another (110) (100) dendrite grown at a higher speed. A large bulge is evident on this specimen, again resulting from rabling the oxide at the seed-blank interface. Several stray grains are apparent in the lower section and in the right side of the bulge. Otherwise, the section is a single crystal. Again,
Figure IV-9: \(<110\>\frac{1}{2}110\frac{3}{2}\) dendrite at 1-1/2 inches from the seed (magnification 12X).

Figure IV-10: \(<110\>\frac{1}{2}100\frac{3}{2}\) dendrite grown at high speed (77 cm/hr) (magnification 12X).
the effect of the growth conditions on the structure is obvious. In
the lower part horizontal \(\{100\}\) sheets developed, and extend over
large distances. At the bottom of the bulge, where growth proceeded
upward, two other sets of \(\{100\}\) sheets (perpendicular to those
developed in the lower section) are evident. Again, at the very top the
horizontal \(\{100\}\) sheets are the "primary" growth forms. In the lower
part of the top region, the horizontal plates are broken, while at the
very top they are almost unbroken. This can be explained as follows.
In the middle section, two sets of \(\{100\}\) sheets are growing simultane-
eously. A cross type of structure would be seen in a perpendicular cut
through this region. Cross type structures are related to the develop-
ment of independent dendrite elements. Thus, at the top of the middle
section, growth proceeding as horizontal \(\{100\}\) sheets from the top of
several mismatched vertical elements will produce mismatched horizontal
sheets. Eventually one of these elements suppresses the rest during
their further extension upward and unbroken sheets are evident near the
top. (These may become independent "line" type elements as growth
proceeds along the axis of the crystal.)

3. \(\{\overline{1}1\overline{1}\}\) Dendrites

Figure IV-11 shows a \(\{\overline{1}1\overline{1}\}\) dendrite grown from a \(\{\overline{1}1\overline{1}\}\ \{\overline{1}1\overline{0}\} \{\overline{1}1\overline{2}\}\)
seed. The section is near the junction between seed and casting.
Several stray grains are evident at the right in the bulge at the top.
No \(\{100\}\) plates are particularly well developed. The \(\{\overline{1}1\overline{1}\}\) dendrite
is completely suppressed by stray grains at 1-1/2 inches from the seed.
The experiment was repeated with a similar result.
Figure IV-11: $\langle 111 \rangle$, $\langle 110 \rangle$, $\langle 112 \rangle$ dendrite with strays at top and lower right (magnification 12X).
D. Discussion of Results

Several comparisons can readily be made of the structures observed for the extreme orientation $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$. First of all, it seems clear that $\langle 100 \rangle$ dendrite planes develop extensively when they are aligned nearly in the heat flow direction. In $\langle 100 \rangle$ dendrites there are two such planes, in $\langle 110 \rangle$ one, and in $\langle 111 \rangle$ there are none.

Although no experiments were carried out relating specifically to the development of preferred orientation, it is clear that dendrites decrease in stability from $\langle 100 \rangle$ to $\langle 110 \rangle$ to $\langle 111 \rangle$ when in competition with a random assortment of stray grains. It is of interest, however, to note the relative stability of a $\langle 110 \rangle$ dendrite, which although not as stable as a $\langle 100 \rangle$, is far more stable than a $\langle 111 \rangle$ dendrite. The extensive development of a $\langle 100 \rangle$ dendrite sheet coincides with stability of a dendrite. In Chapter III it was suggested that the enhanced heat transfer in a fully developed sheet stabilizes its growth. The difference between $\langle 110 \rangle$ and $\langle 111 \rangle$ orientations would be explained by this effect. The smaller difference between $\langle 110 \rangle$ and $\langle 100 \rangle$ might also be explained since two sheets are better than one. (Differences in interface kinetics, secondary growth forms, and/or solute rejection might also be significant.) Although no direct evidence exists for the mechanism of suppression of one grain by another, the extension of a $\langle 100 \rangle$ dendrite sheet sideways suggests itself in the case of $\langle 100 \rangle$ dendrites, and in the case of $\langle 110 \rangle$ dendrites in one direction. A terminal instability discussed in Chapter III might explain why a dendrite element stops growing at the boundary region between two grains, allowing the side extension of a neighboring dendrite sheet.
The factors determining dendrite growth form spacing and order of branching have not been studied in detail. Several features were noted, however. Under the conditions of the experiments, little branching transverse to heat flow was evident at low growth rates (high heat though not relative to heat of fusion), while secondary branching did occur transversely at high growth rates. Also, in several crystals only primary close packed dendrites occurred near the top and primary dendrites with vertical secondary branches at the bottom. Perhaps natural convection in the liquid is responsible for this difference. In Chapter III, the primary dendrite spacings of some of these crystals were plotted versus solidification time on the same graph as dendrite spacings from columnar dendrites (see Figure III-13). Primary spacing depends little on solidification time in these crystals, as discussed in Chapter III. The reason suggested for the relative stability of primary dendrite crosses or sheets under high heat flux was the reduction in the effectiveness of "terminal instability". With a high gradient, a dendrite would not tend to fall far behind its neighbors. If a relatively finer primary spacing were stable, then less secondary branching would be required to satisfy the side diffusion requirements of the thickening sheets. As a result, the primary sheets would be thicker, as noted, and less branching would occur. Needless to say, this is only a qualitative explanation and does not account for the absence of transverse branching, or the close packed dendrite structure at the top of the several crystals (perhaps convection affects the local thermal gradients.) Further work is necessary to establish these ideas or others in order to account for the complex experimental results.
V. DOUBLE ORIENTATION

Introduction

Under conditions of unidirectional growth a strong preferred orientation develops in a casting or an ingot. Typically, this orientation corresponds to all grains having a $\{100\}$ plane within $10^\circ$ of the chill surface (after growth of several inches from the chill surface). However, the orientation of grains with respect to a perpendicular to the chill surface is random.

In a study of the application of preferred casting orientation to magnetic materials, Fisher and Walter have made seeded ingots of silicon-iron in which all crystal have the $\{100\} [001]$ orientation with respect to the sides of the ingot. Two types of seeds were used. The first type was a section of a previously cast unidirectional ingot, which had a preferred orientation along one axis (Fisher and Walter call these singly oriented ingots). "Seed slabs were cut from these ingots such that the seeding surface was parallel to the columnar axis of the slab, and to the $\langle 001 \rangle$ direction of the crystals. The slabs were placed at the base of the mold so that the columnar axes of the crystals were perpendicular to the long axis of the mold and parallel to one edge of the mold." See Figure V-1. The ingots made using these seeds "... developed columnar structures but the degree of double orientation of the crystals, as determined by X-ray diffraction and visual observation, was considerably less than perfect." The second type of seed, which was very effective, was made up of laminated strips of commercial grain oriented Si-Fe. The results, presented in a pole figure, showed most grains within $10^\circ$ of being doubly oriented. The grains which were outside this limit apparently did not grow from the seed plate.
Figure V-1: Schematic diagram of solidification of a doubly-oriented ingot from a singly-oriented ingot (see an editor).

Figure V-2: Sketch showing mold configuration of casting in which doubly oriented dendrites were produced (Polich, et al.). Chill has hatch marks.
Polich et al.\textsuperscript{56} have succeeded in growing doubly oriented grains in steel by growing grains around a corner. See Figure V-2. In this case single orientation develops during perpendicular growth from the chill. On reaching the corner in the mold, the heat flow direction gradually changes. Dendrites with a $\langle 100 \rangle$ direction in the new heat flow direction are soon favored over other singly oriented grains. Thus, grains left growing vertically in Figure V-2 are doubly oriented. The principle is very similar to Fisher and Walter's seed slabs, except that seeding is avoided.

**Change in Growth Direction - Effect of Orientation**

Consider the details of the change in growth conditions during growth around a corner. Two grains are shown in Figure V-3a. Both are oriented with a perfect "single orientation"; namely, with a $\langle 100 \rangle$ direction perpendicular to the chill. In a cross-section perpendicular to heat flow, however, their orientation differs by 45° (see Figure V-3b). As the heat flow direction changes gradually during growth around the corner, different crystallographic directions will become aligned with the heat flow. This is shown schematically in the stereographic projections of Figure V-3c; the arrows indicate how the direction aligned with heat flow changes. Grain 1 will pass from a [100] growth direction to a [001] growth direction (along the [010] zone). Notice that one dendritic plane is always aligned parallel to heat flow during the change in heat flow direction. Compare the sketch of Grain 1 in Figure V-3 to the actual structure of a $\langle 100 \rangle$ grain observed in the boat furnace experiment (Figure IV-3, Chapter IV). Notice that Grain 1 is oriented at most at 45° to the heat flow direction (when heat flow is its [101] direction). It is interesting to recall from Chapter IV that a $\langle 110 \rangle$ dendrite is not easily suppressed by its neighbors. Grain 2, on the other
Figure V-3: Schematic diagram of two grains in a double orientation mold. Grain 1 is oriented with $\langle 100 \rangle$ directions in both the original and final growth directions, perfect for double orientation. Grain 2 is oriented with a $\langle 100 \rangle$ direction in the first growth direction, and a $\langle 110 \rangle$ direction in the final growth direction. Grain 2 is oriented perfectly for single orientation, badly for double orientation. The initial growth direction is $\langle 100 \rangle$, the final growth direction is upward; the path of change in heat flow direction is indicated by arrows on the stereographic projections.
hand, changes from the [100] growth direction to the [011] direction (along the [011] zone). Although the final [011] direction is not very unfavorable for dendritic growth, the dendrite must grow for a time with a [111] direction aligned with the heat flow. <111> dendrites, as shown in the previous chapter, are quickly suppressed. It would seem that a grain oriented so as to produce double orientation (as Grain 1) should survive the change in heat flow direction with relative ease, while a grain which was not doubly oriented would be suppressed, as is Grain 2. Naturally there are intermediate cases between these two examples.

**Experiments Performed**

The introduction of a second chill in the vertical direction seemed reasonable in order to provide a quick change in heat flow direction and sufficient gradients during growth in the vertical section. The mold designed to accomplish this is shown in Figure V-4. The drag mold material was CO₂ sand preheated to 300°C, the cope section calemo refractory preheated to 700°C. The chills were water cooled.

The expected grain structure of a section through the casting is shown in Figure V-5A. Single orientation is achieved by growth from Chill 1. At the same time, solidification proceeds from a second chill (Chill 2). By the time dendrites growing from Chill 1 reach the main plate section, the cooling front from Chill 2 will reach the junction. The dendrites from Chill 2 arrive at the junction slightly after those from Chill 1, so it is Chill 1 dendrites that grow up into the plate.
Figure V-4: Two chill double orientation mold.
A bottom chilled 3/4 inch thick by 4 inches wide by 8 inches high plate casting was made, so that dendrites produced with a single chill (singly oriented) could be compared to doubly oriented dendrites. The cope and drag preheat temperatures in the single chill mold were the same as in the double orientation mold. In both molds, aluminum-4.5 per cent copper was poured at 700°C and the water turned on through the chills immediately before pouring.

Results

Figure V-5B shows the vertical structure of the doubly oriented casting. Note that the structure is very similar to that desired (Figure V-5A) except that some grains from Chill 2 grew past the junction. Figure V-6 shows the dendrite structure (perpendicular to heat flow) at 1-5/8 inches from the top of the junction in the doubly oriented casting. Figure V-7 shows the corresponding structure in the singly oriented casting taken at 3-1/2 inches from the chill.

Extent of Doubly Orientation

As discussed earlier, transverse sections through columnar dendrites often show a cross-type structure. If orientation differences relative to the final heat flow direction are neglected, extent of double orientation can be determined by the orientations of dendrite crosses. A dendrite cross has four-fold symmetry. Therefore any description of its orientation beyond 90° is redundant (it corresponds to orientations in the first 90°).

If a perfectly doubly oriented grain is oriented as follows:
Figure V-5: Two chill double orientation mold casting. (a) Grain structure expected. (b) Actual structure, poured at 700°C.
Figure V-6: Dendrite structure perpendicular to heat flow in doubly oriented casting (representing about 6 per cent of total area of cross section, all of which was measured, magnification 12X).

Figure V-7: Dendrite structure perpendicular to heat flow in singly oriented casting (section similar to above).
Then a perfectly oriented dendrite element is as follows and a totally imperfect dendrite is mis-oriented by 45°. A neutrally oriented dendrite is half way between, namely 22.5° to either side of the perfect dendrite.

Define a pair of neutral lines inclined 22.5° from perfect orientation.

The angle between either neutral line and perfect is defined as positive and may vary from 0 to 22.5°. That between a neutral line and the 45° degree line on its respective side is defined as negative, from 0 to -22.5° on each side.

Thus each grain or subregion may be assigned one θ value, since it can have only one symmetry element (dendrite arm) between the 45° lines where angles are defined. Each grain or subregion also has an area A of intersection in the cross-section under consideration. θ_average (θ_{av}) is a measure of the average angular displacement from the neutral line:

\[ \theta_{av} = \sum_{i=1}^{n} \frac{A_i \theta_i}{A_{total}} \]

where \( n \) is the number of grains or subregions, whichever are measured.

A figure of merit for per cent double orientation (D.O.) is the ratio of average orientation to perfect orientation or
\[
\text{D.O. } = \frac{\theta_{av}}{22.5}
\]

Perfect double orientation is 100 per cent.

Neutral double orientation is 0 per cent.

Negative percentages show a degree of double orientation against theoretical expectations, and probably result from a non-statistical sample.

<table>
<thead>
<tr>
<th>totally imperfect</th>
<th>neutral</th>
<th>perfect</th>
<th>neutral</th>
<th>totally imperfect</th>
</tr>
</thead>
<tbody>
<tr>
<td>×</td>
<td>×</td>
<td>+22.5°</td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>-22.5</td>
<td>0°</td>
<td>+22.5°</td>
<td>0°</td>
<td>-22.5°</td>
</tr>
</tbody>
</table>

Using this figure of merit, the extent of double orientation was determined in the doubly oriented casting and in the control singly oriented casting described above. The doubly oriented casting shows 40 per cent double orientation by this figure of merit. This result is in qualitative agreement with Fisher and Walter for their first seeding arrangement, in which the "degree of double orientation . . . was considerably less than perfect". The singly oriented casting showed 8 per cent double orientation; this compared well with the theoretical value of 0 per cent and contrasts with 40 per cent in the double oriented casting. One hundred and twenty six grains were averaged in the doubly oriented casting, and one hundred and thirty seven grains were averaged in the singly oriented casting. Thus, it seems that this method of determining extent of preferred orientation is statistically significant.
VI. EQUIAXED DENDRITE STRUCTURE

Introduction

Classically, equiaxed grains have been described as those which occur at the termination of the columnar zone, usually in the middle of the casting or ingot. Under some conditions, such as low thermal gradients, the entire ingot may be equiaxed. Equiaxed grains are characterized by their lack of extension in any direction, as the name implies, and by lack of preferred orientation.

In an often quoted paper Winegard and Chalmers\textsuperscript{57} explained the origin of the equiaxed zone in terms of constitutional supercooling which caused new grains to nucleate in front of columnar grains. Recently\textsuperscript{58} Chalmers has pointed out weaknesses in this theory and proposed an alternate hypothesis in which all nuclei are formed during the period of supercooling before growth. In the discussion of the chill zone in Chapter II, the effect of convection in causing nucleation of grains was mentioned. Such convection constitutes a third possible mechanism for formation of the equiaxed zone in ingots.

Reason for Study

Equiaxed grains were studied briefly for two reasons. One reason was to gain information directly on dendrite morphology, thus complementing the work on columnar dendrites. Northcott\textsuperscript{27} and others have showed the general similarity in etched sections between columnar grains and equiaxed grains. In this work sections of this type were examined, and decanting experiments also performed. Normal decanting of columnar structures is difficult, due
the liquid layer left adhering to the solid, especially in recesses. A new method of decanting described below, although not very effective with columnar dendrites, worked well with equiaxed dendrite, and was useful in showing their structure.

Second, equiaxed grains were made to study segregation during freezing, as described in the next chapter. In this case, decanting provided a method for interrupting solidification.

**Apparatus**

The apparatus used was a modification of the equipment used by Brody\textsuperscript{15} in his study of slowly solidified dendrites. See Figure VI-1. The mold used was a steel shell lined with Fiberfrax paper. The mold was held in a preheated furnace identical to that used in making columnar ingots (Chapter III). Molten aluminum was introduced into the mold through a stainless steel funnel which fed into a coated iron pipe; the mold was filled about half way. Three inches from the bottom of the mold there was a fiber-glass screen held in place by Fiberfrax cement. This screen, closely fitting the iron pipe and a thermocouple which penetrated it, enclosed most of the liquid aluminum. The thermocouple in the mold was attached to another thermocouple in the furnace so as to produce a differential output. By maintaining this output constant, a relatively constant rate of heat extraction was obtained. Each run was started with metal and mold at 700°C, then differential control was set so that the ingot would freeze very slowly (see Table VI-1).

While the aluminum cooled and started to solidify, a crucible of lead was melted nearby and superheated to about 700°C. The lead was removed from the furnace and stirred. A second thermocouple in the aluminum was
Figure VI-1: Diagram of apparatus used in lead decanting experiments. Solidification controlled at constant rate of heat extraction by means of differential thermocouple.
attached differentially with a thermocouple in the lead. When the temperature difference between the two had dropped to the order of 10°C, the lead was poured into the funnel at the top of the furnace. The lead ran through the iron pipe and out into the bottom of the mold, displacing aluminum. Enough lead was used to displace all liquid aluminum below the screen. Solid dendrites, however, although tending to float on the lead, were caught by the fiber-glass screen (see Figure VI-2). Thus the liquid and solid aluminum were separated. At this point the furnace was turned off and the door opened.

Results

Two experiments of the above type are discussed here (see Table VI-1).

<table>
<thead>
<tr>
<th>Freezing Time from Liquidus to Lead Pour</th>
<th>Estimated Total Solidification Time</th>
<th>% Cu in Liquidus to Eutectic Alloy</th>
<th>Lead Pouring Temp.</th>
<th>Approximate Solid at Pouring Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lead Decanting Experiment 1</td>
<td>3 hours</td>
<td>10 hours</td>
<td>3.95</td>
<td>1187</td>
</tr>
<tr>
<td>Lead Decanting Experiment 2</td>
<td>4 hours</td>
<td>100 hours</td>
<td>4.51</td>
<td>1201</td>
</tr>
</tbody>
</table>

After cooling to room temperature and removal from the mold, the ingots were reheated to 350°C (above the melting point of lead). Shaking of the ingot was necessary to remove the last traces of lead. In cases where only a small amount of aluminum alloy had been allowed to solidify before the lead was added, the ingot (during lead re-melting) was held in a fiberglass sack which allowed the lead to run through to a crucible (also, the sack was shaken).
Figure VI-2: Schematic sketch of lead decanting. Before pouring lead solid aluminum (dendrites) form in the liquid melt. After lead is poured, liquid aluminum is displaced by liquid lead at the same temperature, leaving the dendrites trapped by a fiberglass screen.
The structure of dendrites from Lead Experiment 1 is shown in Figure VI-3. The structure shows sheet-like dendrite crosses, and clearly distinct dendrite elements within a single grain. It is remarkably similar to the structure of columnar dendrites, except that more than one growth direction may develop in one grain.

Segregation measurements on lead decanted dendrites are discussed in the next chapter.
Figure VI-3: Equiaxed dendrites from lead decanting experiment 1. Many dendrite elements can be seen in a single grain or dendrite. Each is made of two intersecting (primary) sheets tapering to a point at their intersection, which is the dendrite growth direction. The structure resembles an array of Phillips head screwdrivers. Higher order dendrite branching is also evident.
VII. DENDRITIC SEGREGATION

Introduction

In the general introduction of Chapter I, some mention was made of solute segregation expected as a result of dendritic growth. Two very different types of segregation exist and must be distinguished. The first type is macrosegregation. Macrosegregation is the change in average chemical composition of one or more components that occurs over relatively large distances in a casting or ingot. Inverse segregation, settling, and "V" and "inverse V" segregation are examples of this type of segregation. Macrosegregation has been studied extensively by various workers, normally employing chemical analysis of drilled samples. Only in the case of inverse segregation has quantitative comparison been made between theory and experimental results.

Segregation resulting from normal freezing can be considered an intermediate case between macrosegregation and microsegregation. It is like macrosegregation because it produces segregation of components "over relatively large distances in a casting or ingot". However, the alloy composition resulting from normal freezing is locally homogeneous.

Normal freezing is described by the same equation, \( C_S = k C_0 (1 - f_0)^{k-1} \), that was mentioned in the General Introduction of Chapter II for dendritic freezing. In fact, dendritic freezing may be considered as normal freezing (with \( K_{\text{eff}} = K \)) occurring over volumes with dimensions of the order of dendrite spacings.\(^{44}\)
Study of the segregation that results from dendritic freezing (microsegregation) has been limited by experimental techniques. The main difficulty is a simple one: the distance between the maximum and minimum solute contents, which define the normal freezing elements, are very small, often 1/10 millimeter or less. Several early investigators attempted to determine solute concentration profiles in these regions by drilling out samples and making gradients. Brenner and Kostorn \(^{59}\) conducted an investigation on microsegregation using micro-hardness measurements, and found significant concentration gradients. As mentioned in Chapter I, Michael and Bever \(^{14}\) found a minimum copper content of 2.0 per cent Cu in a 4.06 per cent Cu casting. The discrepancy between this value and the predicted .6 per cent was not rigorously examined, although diffusion in the solid was considered a possible explanation.

**Experimental Technique**

A recent development in X-ray technique, electron beam micro-analyses, has been used in this study to determine the segregation patterns existing in dendrites. The specimen to be investigated was mounted in bakelite, polished to fine diamond, and lightly etched. The dendrite element to be studied was marked with microhardness indentations, and a photograph taken. The specimen was then carefully polished once again to fine diamond.

In the microanalyzer high energy electrons are magnetically focused onto a small spot on the specimen (about 1 micron in diameter). The spot position is set with the aid of the microhardness indentations and a photograph of the structure. In the case of aluminum-copper alloy used in this investigation 15KV electrons excite the CuK\(_{\alpha}\) characteristic radiation.
Diffraction from a LiF crystal eliminates other wavelength radiation, and the Kα intensity is determined using a scintillation counter with a pulse height analyzer. The ARL unit had a take-off angle of 15.5°.

**Standardization**

A pure copper target gave I₀ uncorrected for copper Kα. A pure nickel target gave the background radiation of wavelength CuKα (nickel is used because it is next to copper in atomic weight); uncorrected I₀ from copper minus nickel background gives I₀. The background intensity was also measured for pure aluminum and the value subtracted from the intensity recorded for the aluminum-low copper alloys, giving I. Ziebold has made theoretical calculations showing an approximate 8 per cent negative deviation from linearity between I/I₀ and fraction copper in the alloy, but Bardes has made standards which lie near a perfect straight line. In this work weight per cent copper is taken as I/I₀, in conformance with Bardes' results. The path of the probe can be seen on the specimen (due to a soot deposit from diffusion pump oil) and becomes broader and darker upon etching lightly (see Figure VII-1).

**Dendrites Studied**

Segregation patterns in several different types of dendrites were determined. The first type were the columnar dendrites discussed in Chapter III. Figure VII-1 shows the probe pattern taken on one specimen from water cooled Columnar Ingot 1, the specimen cut out at 7 inches from the chill surface. Figure VII-1 also shows the composition along the probe path. A comparison of the photomicrograph to the composition variations shows that visual observation of darkness changes correspond closely to concentration
Figure VII-1: Composite figure showing microprobe trace and photograph of microstructure of A-10 chilled Columnar Ingot 1 at 7 inches from the melt. Both graph and photograph have same scale in the horizontal direction.
changes measured by the probe. Specimens from the water cooled ingot taken very near the chill and at 3 inches were also studied.

As shown in Figure VII-1, a minimum in composition exists at the intersection of the primary dendrite sheets. In all cases this probe path minimum was scanned manually to determine the lowest solute content, which might be off the point of intersection of the probe path. Since the concentration gradients in the minimum area were very low, little difference was found between the minimum on and off the probe path in most cases.

Specimens were taken from air-cooled Columnar Ingot 2 very near the chill and at 7 inches from the chill. Due to the large size of dendrites in this case (relative to the probe traversing speed) paths of the type shown in Figure VII-2 were made and the minimum copper content determined.

In order to determine whether or not minimum solute content changed during solidification due to diffusion in the solid, dendrites from air-water cooled Columnar Ingot 4 were examined on the probe. In this ingot dendrites were formed by air cooling until the liquidus was about 3/4 inches from the chill surface, and then water cooling (quenching). Even if diffusion in the solid might occur over shorter diffusion distances during water cooling, it would be much less significant over the distances characteristic of air-cooled dendrite spacings. Probe data were taken on dendrites at 1/2 inch, 3/4 inch, and 7/8 inch.

Another type of interrupted solidification was studied on the probe. The lead decanted equiaxed dendrites discussed in Chapter VI were examined. In both cases, the copper composition was approximately constant across a
Figure VII-2: Photomicrograph showing probe path used to find minimum percent aggregate, coarse dendrites, specimen in section 3 inches from the chill in air chilled Columnar Ingot 2 (magnification, 50X).
solid element except in a microscopic outer layer (probably due to rapid solidification of a thin layer of liquid aluminum not displaced by the lead).

Results

Results of the electron beam microprobe studies on the specimens mentioned above are listed in Table VII-1. The chemical composition of the alloy used in each case was determined by chemical analysis of a chill cast sample and is listed in the Table. Only minimum copper contents in the dendrites are given, since these values can be compared directly. In case of a variation in composition of the alloy from 4.5 per cent copper, minimum compositions were normalized to a 4.5 per cent copper basis for comparison by multiplying the observed per cent copper by 4.5 and dividing by the overall alloy composition.

Discussion of Results

In Table VII-1 it can be seen that the minimum per cent copper in the water-chilled columnar dendrites is about 1.3 to 1.4 per cent copper. The minimum in the air-chilled ingot is 1.4 at 7 inches, but 2.4 per cent near the chill. Brody\textsuperscript{16} predicts that for cooling curves of similar shape, assuming diffusion of solute in the solid, extent of diffusion changes only slowly with solidification time, which is well supported by four of the above five values. The 2.4 per cent value may be due to the nature of the cooling curve for air cooling (see Figure III-11) in which cooling rate decreased with time. This allows additional time for solid diffusion later in solidification relative to the dendrite spacing established early in solidification.
In the air-water columnar ingot, a value of .83 per cent copper was obtained as a normalized minimum at 1/2 inch from the chill. The value is close to the equilibrium minimum per cent copper expected; when compared to the 2.4 per cent minimum found in a fully air-cooled ingot, it proves that substantial homogenization occurred during subsequent freezing and cooling in the later case.

The same effect is observed in the equiaxed grains. At 5 per cent solid, the minimum per cent copper is in virtually perfect agreement with the phase diagram, while after 40 per cent has been allowed to solidify, the minimum increased to .98 per cent copper (normalized). After complete solidification of the same alloy at the same cooling rate Brody found a minimum of about 2 per cent copper in the center of the dendrites, in agreement with the results of Michael and Bever. The conclusion is that diffusion must occur in the already solid portion of dendrites during later solidification, both for columnar and equiaxed dendrites. Brody's computer calculations are in good quantitative agreement with the results of Table VIII-1.
TABLE VII-1

Electron Beam Microprobe Analyses

<table>
<thead>
<tr>
<th>Unidirectional Ingot</th>
<th>Sample % Copper</th>
<th>Minimum % Copper Probe</th>
<th>Normalized Minimum % Copper</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air Chilled Columnar Ingot 1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>chill</td>
<td>4.81</td>
<td>2.4</td>
<td>2.24</td>
</tr>
<tr>
<td>7 inches</td>
<td>4.81</td>
<td>1.41</td>
<td>1.32</td>
</tr>
<tr>
<td>Water Chilled Columnar Ingot 2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>chill</td>
<td>4.65</td>
<td>1.43</td>
<td>1.39</td>
</tr>
<tr>
<td>3 inches</td>
<td>4.65</td>
<td>1.48</td>
<td>1.43</td>
</tr>
<tr>
<td>7 inches</td>
<td>4.65</td>
<td>1.33</td>
<td>1.29</td>
</tr>
<tr>
<td>Air, then Water Chilled Columnar Ingot 4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1/2 inch</td>
<td>5.05</td>
<td>.94</td>
<td>.83</td>
</tr>
<tr>
<td>3/4 inch</td>
<td>5.05</td>
<td>1.0</td>
<td>.89</td>
</tr>
<tr>
<td>7/8 inch</td>
<td>5.05</td>
<td>1.58</td>
<td>1.40</td>
</tr>
</tbody>
</table>

Equiaxed Grains - Lead Displacement

<table>
<thead>
<tr>
<th></th>
<th>Minimum % Copper</th>
<th>Normalized Minimum % Copper</th>
</tr>
</thead>
<tbody>
<tr>
<td>1187°F Interruption (40% Solid)</td>
<td>3.95</td>
<td>.86</td>
</tr>
<tr>
<td>1201°F Interruption (5% Solid)</td>
<td>4.51</td>
<td>.65</td>
</tr>
</tbody>
</table>

Note: Accuracy of probe results is significant to no more than two places.
VIII. SUMMARY AND CONCLUSIONS

1. A study was made of dendritic structure at the chill surface. Grain sizes at the chill surface can be quite large (3 centimeters diameter) even at rapid cooling rates, provided there is minimum convection after solidification is initiated, and no grain refiner is employed. Surface dendrite traces (dendrite "arms") are not of $\langle 100 \rangle$ orientation. They are either the intersection of $\langle 100 \rangle$ dendrite sheets with the chill surface or projections of $\langle 100 \rangle$ directions onto the chill surface. The $\langle 100 \rangle$ sheets extend from the chill surface into the casting while projections of $\langle 100 \rangle$ directions exist only at the chill surface.

2. Dendritic growth perpendicular to heat flow in the chill region generally occurs by simultaneous operation of two mechanisms. One involves growth on the chill surface and other involves growth above the chill surface. Two types of growth on the chill surface were described in the preceding conclusion. The intersection with the surface of $\langle 100 \rangle$ dendrite sheets extend along the surface. In a given dendrite the sheets which grow in this way are those most nearly perpendicular to the chill. Dendrite "arms" also grow along the surface in directions which constitute the projections of $\langle 100 \rangle$ directions onto the chill surface. Growth above the chill surface, on the other hand, does not involve the chill surface itself. In this case $\langle 100 \rangle$ dendrite sheets develop which lie at a low angle to the chill surface. These grow extensively in a plane that is more or less parallel to the chill surface.
3. Bent dendrite traces on the chill surface are frequently observed when conditions of strong convection exist during initiation of solidification. Bending is also frequently observed in dendrites which grow in the absence of convection. Although less severe, bending in the latter case may cause an orientation change of 20 or more degrees and an apparent radius of curvature of less than one centimeter in the chill surface. Bending is such that the crystal lattice is concave to the chill surface.

4. The orientation of surface grains is random. There is slight, if any, tendency for grains to be larger the more nearly a \( \langle 100 \rangle \) direction approaches perpendicularity to the chill.

5. Columnar ingots of aluminum-4.5 per cent copper alloy were produced by unidirectional solidification using air and water as coolants. The dendrite structure of these columnar ingots consists of essentially sheet-like dendrite growth forms, both for rapid and slower cooling rates. Well developed primary, secondary and tertiary dendrite growth forms were identified. Dendritic structure in a longitudinal section (parallel to heat flow) differs markedly from the structure in a transverse section (perpendicular to heat flow). In the longitudinal sections there are long narrow dendrite traces (primary "arms") nearly in the heat flow direction, wider "band" intersections at various angles to the primary "arms", and secondary arms nearly perpendicular to heat flow. In a transverse section, the primary structure consists of a pattern of dendrite "crosses" or "lines" within a single dendrite.
The difference in structure between longitudinal and transverse sections is due to the existence of a \( \langle 100 \rangle \) preferred orientation in columnar dendrites, and also due to the fact that only those \( \{100\} \) dendrite planes at a high angle to the chill surface develop extensively.

6. Columnar dendrites tend to form independent dendrite elements during growth. Dendrite element size approaches primary growth form spacing during growth away from the chill surface. The boundaries between dendrite elements are regions of high solute content, often containing second phase. Misorientations exist between dendrite elements. A mechanism is proposed for dendrite element formation. The existence of dendrite elements helps explain the development of a preferred orientation.

7. Single crystal dendrites of simple orientations have been prepared in a crystal growing furnace of Chalmers' design. Transverse sections through \( \langle 100 \rangle \) dendrites have a cross-type structure, similar to most columnar dendrites. \( \langle 110 \rangle \) dendrites have a "line" type structure, corresponding to the development of \( \{100\} \) dendrite planes which lie perpendicular to heat flow. These are more perfectly developed if they are vertical during growth than if they are horizontal during growth. \( \langle 111 \rangle \) dendrites have a poorly developed (discontinuous) dendrite growth form structure and are quickly suppressed by stray crystals. Stray crystals are also found in the \( \langle 100 \rangle \) and \( \langle 110 \rangle \) dendrites. Although neither \( \langle 100 \rangle \) nor \( \langle 110 \rangle \) are easily suppressed by strays, the strays grow larger in competition with \( \langle 110 \rangle \) then in competition with \( \langle 100 \rangle \).
8. Dendrite growth form spacing has been measured as a function of solidification time. For primary growth form spacing in water chilled columnar ingots, spacing is proportional to the square root of solidification time. This result is not achieved in air chilled columnar dendrites and seeded dendrites. In these cases, primary growth form spacing is much less dependent on solidification time. Secondary and tertiary growth form spacings are usually nearly equal, especially on a logarithmic scale. Plotted together their spacing was found to be proportional to about the fourth root of solidification time.

9. Equiaxed dendrites were produced by slow solidification. Solidification was interrupted by decanting with molten lead. The structure produced was similar to columnar dendrites in many ways. \(<100\) dendritic planes were developed and dendrite elements were in evidence. The main difference is that growth occurred in all directions, so that groups of dendrite elements developed oriented in all three \(<100\) directions.

10. Doubly oriented ingots were produced using two chills; the structure was compared to normal columnar grains, which are singly oriented (having one \(<100\) direction aligned preferentially). A figure of merit was derived for double orientation which can range from minus 100 per cent to plus 100 per cent. Zero per cent represents no double orientation, 100 per cent is perfect double orientation and minus 100 per cent is perfect double orientation, but contrary to theoretical prediction. A successfully doubly
oriented ingot was 40 per cent doubly oriented by this figure of merit. A control singly oriented casting was 8 per cent doubly oriented.

11. The electron beam microprobe was used to determine segregation of copper in columnar and equiaxed grains of aluminum-copper alloy. Segregation in grains produced by interrupted solidification (lead decanting or water quenching of air cooled structures) was compared to segregation in grains that had solidified without interruption. The results showed that the first solid to solidify has a composition which is predicted from the phase diagram, and that the minimum composition increased during solidification due to solid state diffusion. Quantitative results were given.
IX. SUGGESTIONS FOR FURTHER WORK

1. An extension of the present work on aluminum to other alloy systems would have several goals: first, the determination of dendrite morphology in other cubic systems, both FCC and BCC; second, the determination of dendrite structure in metals having non-cubic symmetry, particularly hexagonal metals. In hexagonal metals, it is worthwhile to re-examine the preferred orientation at the chill surface and in the columnar region. If there is a strong growth anisotropy on the chill surface in hexagonal metals, as previously reported\textsuperscript{20a}, it would be of interest of measure it quantitatively and compare various systems.

2. The mechanism of formation of equiaxed grains is of fundamental interest in the study of casting and ingot structure. Nucleation of grains by convection should be studied as a possible explanation of many ingot structures. Controlled conditions of no convection and forced convection would be studied.

3. Bending of dendrites needs further study. The "bending" ranges from very severe bending in convected grains to less severe bending of large chill grains to low angle misorientation of columnar dendrites. The field of study might range from "deformation" studies during convection to dislocation studies in columnar grains. The mechanisms of misorientation production must be clarified.

4. More sorting of the parameters affecting primary and higher order dendrite spacings is necessary. The parameters include solidification time, alloy composition, phase diagram characteristics, thermal gradients,
solid-liquid interface structure and the dendrite structure. Dendrite structure affecting dendrite spacing seems paradoxical, but when an element of liquid cools it will not freeze unless there is solid nearby. The form and distribution of this solid must influence the distribution of solid that forms in the element. In this regard, the effect of thermal gradients in the liquid on primary dendrite growth form spacing needs further study. The primary growth form spacing will affect both the structure of secondary growth forms and the size of dendrite elements.

5. Study of the dendrite tip geometry by lead decanting might be useful. Diffusion and heat transfer solutions giving dendrite growth velocities depend on geometry. Also, the form of the tip might elucidate the mechanism that determines dendrite growth direction. The growth of projections of $\langle 100 \rangle$ dendrite directions on a chill surface would be understood more clearly. The same experiments could reveal details of dendrite sheet formation and dendrite element formation, both of which require further study for complete understanding.

6. The development of preferred orientation in columnar growth needs further careful and detailed study. The experimental technique should employ bicrystals like those used by Chalmers and co-workers. Crystals of various orientations could be grown under conditions where their dendrite sheets converge, diverge and run parallel, at various angles of inclination. The structural details of encroachment would be studied.

7. In slowly solidified equiaxed grains thermal gradients are very low and are not aligned with the dendrite growth forms. Therefore, thermal gradients are not responsible for the structures which are obtained.
It would be of interest to make a closer study of primary dendrite growth form spacing and details of dendrite element structure under these conditions and compare them to corresponding structures in dendrites produced in a thermal gradient.
REFERENCES


2. Miksch, E., Research Staff, Department of Metallurgy, Massachusetts Institute of Technology, Private Communication.

3. Flemings, M. C., Professor of Metallurgy, Massachusetts Institute of Technology, Private Communication.


13a. Reference 12, pp. 80-81.

13b. Reference 12, p. 78.


34. Chalmers, B., Reference 19, p. 119.


47. Bardes, R., Research Staff, Department of Metallurgy, Massachusetts Institute of Technology, Private Communication.


49. Sutton, J., Freshman Seminar, Massachusetts Institute of Technology, Private Communication.

50. Reference 19, p. 164.


55b. Reference 54, p. 1275.


60. Ziebold, T., Research Staff, Department of Metallurgy, Massachusetts Institute of Technology, Personal Communication.

61. Brody, H. D., Research Staff, Department of Metallurgy, Massachusetts Institute of Technology, Private Communication.


BIOGRAPHICAL NOTE

Terry Frederick Bower was born March 13, 1939 in Stamford, Connecticut, received his secondary school education at Darien High School, Darien, Connecticut, and entered M.I.T. in September, 1956. He received a Bachelor of Science degree in Metallurgy in June, 1960. As an undergraduate, the author received a Freshman Competitive Scholarship, Foundry Educational Foundation Scholarship, and American Society for Metals Scholarship. In September, 1960 he started graduate study at M.I.T., receiving a Master of Science degree in Metallurgy in June, 1962. After working for a short period in the Solidification Laboratory, M.I.T., for the Division of Sponsored Research, the author began work on a Doctorate in September, 1962.

The author was a laboratory instructor in Engineering Materials in the Fall term of 1961 and a recitation instructor in the same course in the Fall term of 1962.

The author is a member of Sigma Xi, the American Foundrymen's Society, the American Society for Metals, and the American Institute of Mining, Metallurgical and Petroleum Engineers.
APPENDIX A

Analysis of the Angles Between the Respective Intersections of a Plane (hkl) with the Three \{100\} Planes of a Dendrite

The three \{100\} planes form three lines of intersection with a plane (hkl). The angles between these lines of intersection should be those observed between dendrite traces on the chill surface if the dendrite traces are intersections of \{100\} dendrite sheets.

(1) The pole of the plane (hkl) (lying in the chill surface) may be found by a Laue back reflection photograph.

(2) The dendrite traces on the surface are assumed to be the intersection of the \{100\} dendrite planes with the (hkl) plane. Therefore, the cross product of (hkl) with 100, 010, and 001 should give the indices of these traces.

(3) The angle between any two traces may be found by carrying out a dot product between them.

**Notation**

(1) Plane (hkl) is \( \mathbf{C} = h\mathbf{u} + k\mathbf{v} + l\mathbf{w} \)

(2) Dendrite planes 100, 010, 001 are \( \mathbf{P}_{100} = \mathbf{u} \), \( \mathbf{P}_{010} = \mathbf{v} \), \( \mathbf{P}_{001} = \mathbf{w} \)

(3) Dendrite traces are \( \mathbf{T}_1 \), \( \mathbf{T}_2 \) and \( \mathbf{T}_3 \)
(4) Angle between \( \mathbf{T_1} \) and \( \mathbf{T_3} \) is \( \lambda \), between \( \mathbf{T_1} \) and \( \mathbf{T_2} \) is \( \beta \) and between \( \mathbf{T_2} \) and \( \mathbf{T_3} \) is \( \gamma \).

Results

\[
\begin{align*}
\mathbf{T_1} &= \mathbf{C} \times \mathbf{P_{100}} = 1\mathbf{i} - k\mathbf{w} \\
\mathbf{T_2} &= \mathbf{C} \times \mathbf{P_{010}} = -l\mathbf{u} + h\mathbf{w} \\
\mathbf{T_3} &= \mathbf{C} \times \mathbf{P_{001}} = k\mathbf{u} - k\mathbf{v} \\
\mathbf{T_1} \cdot \mathbf{T_2} &= -hk = \sqrt{1^2 + h^2 - \sqrt{1^2 + k^2}} \cos \beta,
\end{align*}
\]

\[
\cos \lambda = \frac{hk}{\sqrt{1^2 + k^2} \sqrt{1^2 + h^2}} \quad \quad \cos \beta = \frac{hl}{\sqrt{1^2 + h^2} \sqrt{1^2 + k^2}}
\]

\[
\cos \gamma = \frac{lk}{\sqrt{1^2 + h^2} \sqrt{h^2 + k^2}}
\]

Values of \( \lambda \), \( \beta \), and \( \gamma \) calculated for some specific planes \( \text{(hkl)} \) are given in Table A-1. Values corresponding to the intersection of the two \{100\} planes making the largest angles with the \( \text{(hkl)} \) plane are underlined. High angle planes are more likely to develop because they are more nearly in the heat flow direction. Figure II-1 shows the traces of the highest angle planes.
<table>
<thead>
<tr>
<th>(hkl)</th>
<th>$\alpha$</th>
<th>$\beta$</th>
<th>$\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>511</td>
<td>46°</td>
<td>46°</td>
<td>88°</td>
</tr>
<tr>
<td>311</td>
<td>48°</td>
<td>48°</td>
<td>84°</td>
</tr>
<tr>
<td>211</td>
<td>51°</td>
<td>51°</td>
<td>78°</td>
</tr>
<tr>
<td>221</td>
<td>71.5°</td>
<td>37°</td>
<td>71.5°</td>
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<tr>
<td>111</td>
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<td>60°</td>
<td>60°</td>
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<td>26°</td>
<td>77°</td>
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<td>521</td>
<td>65°</td>
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<td>86°</td>
</tr>
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<td>531</td>
<td>75°</td>
<td>21°</td>
<td>84°</td>
</tr>
<tr>
<td>123</td>
<td>68°</td>
<td>80°</td>
<td>32°</td>
</tr>
<tr>
<td>210</td>
<td>90°</td>
<td>0°</td>
<td>90°</td>
</tr>
</tbody>
</table>
APPENDIX B

Melting Practice for Chill Casting Experiments

The molds were prepared by polishing chill surfaces to 600 grit paper, coating with amorphous carbon, and assembling with shims and top. The assembly was coated externally with Fiberfrax cement to prevent air leaks and external contact with the melt. The assemblies were preheated to 150°C for two hours before each run.

The metal was previously alloyed aluminum-4.5 per cent copper alloy. About eight pounds of alloy held in a 20-pound crucible was melted in an electric furnace, and superheated to 50°C above the highest planned pouring temperature. The crucible and metal was removed from the furnace to the casting area. The temperature of the melt was measured with an immersion type thermocouple before each casting was made.
APPENDIX C

Discussion of Duplex Grain Results

Several explanations seem possible to account for the results which were presented in Chapter II on duplex grain sizes. Since these explanations depend primarily on the effect of fluid flow in causing path refinement, the first subject of discussion should be the effect of casting conditions on the extent of fluid flow during solidification. In their study of the "fluidity" of cast metals, Taylor, Flemings and co-workers \(^2\) have determined the mechanism of "choking off" of flow in especially designed molds. For an alloy such as aluminum-4.5 per cent copper with a long freezing range, flow was choked off at the tip of the advancing liquid due to the formation of a "mushy" region of liquid plus solid. The first metal to enter the cold mold was the first to lose its superheat and eventually freeze while the liquid "pushing" it from behind entered the preheated mold at a later time, and froze subsequently. The duplex grain size observed in those castings which did not fill the mold may be interpreted in terms of these results. The fine grain structure is present where liquid flow (filling of the mold) and solidification occurred after liquid movement into the casting had stopped. Casting 10 (Figure II-4b) however, shows some rather fine grains near the bottom. The reason is obscure to the author. Also, a few rather coarse grains are often seen near the top of the fine grained region. These may be the first grains to form, and/or there is less convection during filling at the top of flowing metal.
Figure C-1a shows the aluminum-copper phase diagram. In Figure C-1b a hypothetical temperature gradient is shown. The liquid composition in equilibrium with solid along this temperature gradient is given by the liquidus curve of the phase diagram. This composition gradient is shown schematically in Figure C-1c.

Flemings has suggested two mechanisms whereby grains might be nucleated due to convection, under growth conditions with steep thermal gradients. In one case (see Figure C-2a) low solute bulk liquid of composition \( C_0 \) at temperature \( T_L \) is swept between dendrite arms at temperature \( T_1 \), removing liquid of composition \( C_1 \). Diffusion of heat is several orders of magnitude faster than diffusion of solute, so the temperature of the droplet drops to \( T_1 \) while its composition remains \( C_0 \). It is now \( T_L - T_1 \) degrees below its liquidus temperature and nucleation may occur. The new grain formed in this way in the flowing stream would either be trapped or swept out, and the process repeated.

Flemings' second mechanism is similar to the first. In this case, attention is focused on the fate of the liquid of composition \( C_1 \) that was swept out from between the dendrite axes. This droplet (at temperature \( T_1 \)) would be swept into contact with bulk liquid of composition \( C_0 \) and temperature \( T_L \). Again, thermal diffusion would occur first, liquid of composition \( C_0 \) surrounding the droplet would be cooled toward temperature \( T_1 \) (see Figure C-2b). Again, this region would be below its liquidus temperature. The nucleus formed in this case might be stabilized by a temperature decrease in its surroundings.
Figure C-1: Solute and temperature distribution resulting from unidirectional freezing. A. Phase diagram of aluminum-copper showing the variation of liquidus temperature with composition. B. The temperature gradient in unidirectional freezing (schematic). C. Resulting solute gradient in liquid, assuming the liquid is at the liquidus composition at each temperature.
Figure C-2: Three mechanisms for the nucleation of new grains by convection. A. Liquid of composition $C_0$ is swept between dendrite arms. Since thermal diffusion is fast, the temperature drops to $T_1$ while the composition remains fixed. B. Liquid at temperature $T_1$ is swept out and cools liquid of composition $C_0$. C. Superheated liquid is swept back to the dendrite tips where it disconnects a dendrite segment by melting its connection to the rest of the dendrite.
Another possibility of a similar nature, but not involving a nucleation step, is as follows. Liquid at temperature higher than $T_L$ is stirred back of the dendrite tips (see Figure C-2c) and melts part of a dendrite growth form, disconnecting it from the rest of the dendrite. This loose piece is now a new grain, and the process may be repeated.

Finally, a recent theory due to Chalmers$^6$ may be applied to the results of this experiment. Chalmers has proposed that all the grains in an ingot are nucleated during the initial undercooling which occurs when liquid metal comes in contact with the chill. Some of these are stabilized to form chill grains, columnar grains, equiaxed grains trapped in columnar grains and the central equiaxed grains. Under some conditions, many nuclei are remelted. In the view of the author, Chalmer's theory interprets the results of these experiments as follows. The large chill grains are formed in a region of high thermal gradients where many of the nuclei which first form are quickly remelted, leaving only a few that grow. The small chill grains are formed in liquid that has lost its superheat while entering the mold where convection reduced gradients; all nuclei that form in this case are stable and can grow. The weakness in Chalmer's theory as interpreted by the author are (1) a gradual grain size change should be observed, whereas in fact a quite sharp one occurred, (2) there does not seem to be any reason for remelting at the chill interface itself, even with high thermal gradients. (Remelting away from the chill interface is more plausible.
Cooling rate at the chill is affected very little by superheat under the conditions of this experiment (see Appendix D), so that the number of nuclei formed should not account for the vast grain size difference.

There are two features of the duplex grain size castings which have sharp cut-off points. One is the grain size itself. The other is the process of choking-off fluid flow. It seems reasonable that a theory which predicts a relationship between these two is given support by these results. The first three theories listed depend on the effect of fluid flow in forming new grains directly. The fourth does not.
APPENDIX D

Cooling Conditions and Dendrite Bending in Chill Castings

The small plate castings of Chapter II are permanent mold castings. Most of the resistance to heat flow in castings of this type is interface resistance at the mold surface. The heat transfer coefficient of a carbon coated chill is about 0.040 calories/centimeter$^2$ second $^\circ$C. The heat flow per unit area is given by the equation:

\[ \frac{q}{A} = h\Delta T \]

where: $q/A$ is the heat transfer rate per unit area

\[ h \] is the heat transfer coefficient

\[ \Delta T \] is the instantaneous temperature difference between the surface of the solid and the environment.

Notice that the rate of heat transfer does not depend on time. A constant amount of heat is withdrawn in any increment of time. Therefore, a thicker casting takes longer to cool than a thinner one. Only small temperature differences exist, so that preheat is lost before freezing starts. Although the time elapsed from pouring to the start of solidification depends on preheat temperature as well as casting thickness, cooling rate and solidification time depend only on casting thickness.

An approximation of the gradient at the chill surface can be obtained by equating the heat flow through the interface barrier to the heat flow through the metal at the surface:

\[ \frac{q}{A} = h\Delta T = K \left( \frac{dT}{dX} \right)_{X=0} \]
or \((dT/dx)_{x=0} = h/k \Delta T\), where \(K\) is the conductivity. For the conditions used in making castings in Chapter II the gradient calculated from this equation is about 50°C/centimeter. For a plate 1/25 inch thick the difference in temperature from center to outside would be less than 2°C.

Average cooling rate over the solidification range can be estimated as follows:

\[
H = C_p M \Delta T + m \Delta H_f = \frac{q}{A} (\Delta \theta) = h \Delta T_{AV} \Delta \theta
\]

where:
- \(H\) = latent heat removed from the casting per unit area
- \(C_p\) = heat capacity of metal (an average)
- \(M\) = mass of metal per unit area
- \(T\) = temperature drop considered
- \(m\) = mass of metal solidified per unit area
- \(H_f\) = heat of fusion per unit mass (an average)
- \(\Delta \theta\) = time interval considered
- \(\Delta T_{AV}\) = average temperature difference between the casting and the chill

\[
\Delta \theta = \frac{H}{2h \Delta T_{AV}}. \quad \text{The factor of two is introduced because the casting is chilled from both sides.} \quad \frac{\Delta T}{\Delta \theta} \quad \text{is the average cooling rate:}
\]

\[
\frac{\Delta T}{\Delta \theta} = \frac{(2\Delta T)(h)(\Delta T_{AV})}{H} = \frac{2h(\Delta T)(\Delta T_{AV})}{C_p M \Delta T + m \Delta H_f}
\]

\(M = \rho t\) and \(m = f_s t\), when \(f_s\) is the fraction solid, \(t\) is the casting thickness and \(\rho\) is metal density (an average). So

\[
\frac{\Delta T}{\Delta \theta} = \frac{2h \Delta T \Delta T_{AV}}{\rho t (C_p \Delta T + f_s \Delta H_f)} = \frac{k'}{t}, \quad \text{where} \quad k' \quad \text{is a constant}
\]
Therefore, cooling rate is inversely proportional to casting thickness. For the 1/25 inch plate casting, about one half second is required for cooling from the liquidus, 648°C, to the eutectic, 548°C (with a 2°C maximum temperature difference during the process).

**Dendrite Bending**

An outline of a possible mechanism for dendrite bending follows. High thermal gradients cause differential thermal contraction in a partially solidified dendrite. The resulting bend is then frozen into place.

If $r$ is the radius of curvature of bend, $d$ is the half thickness of the bending growth form, and $\alpha$ is the coefficient of thermal expansion,

$$\frac{\Delta a}{a} = \alpha \Delta T$$

$$\Delta T = C d$$

$$\tan \theta = \frac{a}{r} = \frac{\Delta a}{d}$$

then

$$r = \frac{1}{\alpha \frac{a}{C}}$$

$2d$ is the dendrite thickness, $a$ is the section length considered, and $\Delta a$ is the length difference due to temperature change from one side to another. $\alpha$ is about $0.25 \times 10^{-4}$ °C$^{-1}$ for aluminum. To bend a dendrite with a radius of curvature of 1 centimeter requires about 40,000°C/centimeter.

* Average values of various properties are used both for convenience in calculation and because the major source of error is the value of the heat transfer coefficient. Cooling conditions depend on this value, which will vary from casting to casting. All calculations are approximate.
This is compared to $50^\circ$C/centimeter calculated earlier in this appendix. One factor has been overlooked, however. Some undercooling may occur before solidification starts. In this case, very high local temperature gradients might be generated during nucleation and the growth of "primary" dendrite axes. A somewhat higher undercooling would occur in the thinner castings because the cooling rate is higher. Here the bending has a smaller radius of curvature. (Macroscopic thermal gradients in the absence of undercooling, on the other hand, do not depend on casting thickness.)

Several observations on bent dendrite remain unexplained, even qualitatively. First, why do bent dendrites have a fairly smooth curvature? Second, why are they concave to the chill surface? And finally, how is the structure "frozen in" if it is due to differential thermal contraction, since high thermal gradients will be very transient?
APPENDIX E

Calculation of Relative Areas of the Stereographic Triangle in 5° Angle Ranges from the \( \langle 100 \rangle \) Pole and the \( \langle 100 \rangle \) Zone

Angle Ranges from \( \langle 100 \rangle \) Pole

The relative areas in 5° increments from the \( \langle 100 \rangle \) pole may be found by calculating the relative areas between latitudes from the "north pole". Eight stereographic triangles are symmetrically arranged about such a pole. Beyond the 45° latitude a correction must be made, however, because the stereographic triangles are no longer symmetrical about the pole.

The area between the north pole and \( \theta \) degrees from the pole is

\[
A = \int_0^\theta \int_0^{\pi} r \sin \theta \, d\psi \, d\theta
\]

where \( r \) is the radius of the sphere, \( \theta \) is the angle from the north pole, and \( \psi \) is the longitude. The solution is \( A = 2\pi r^2 (1 - \cos \theta) \). The area between latitude \( \theta_1 \) and latitude \( \theta_2 \) is found by subtraction.

From latitude 45° to 54.7° a correction is made to allow for the overlap of stereographic triangles. The area between latitudes 45° and 50° is multiplied by the correction
factor $A_1/A_2$, from $50^\circ$ to $54.7^\circ$ by $A_3/A_4$. $A_1$ and $A_3$ were measured by graphical integration. The resulting relative areas from the $\langle 100 \rangle$ pole are given below.

<table>
<thead>
<tr>
<th>Angle Range</th>
<th>Relative Area</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 - 5°</td>
<td>381</td>
</tr>
<tr>
<td>5 - 10°</td>
<td>1138</td>
</tr>
<tr>
<td>10 - 15°</td>
<td>1888</td>
</tr>
<tr>
<td>15 - 20°</td>
<td>2624</td>
</tr>
<tr>
<td>20 - 25°</td>
<td>3338</td>
</tr>
<tr>
<td>25 - 30°</td>
<td>4028</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Angle Range</th>
<th>Relative Area</th>
</tr>
</thead>
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<tr>
<td>30 - 35°</td>
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</tr>
<tr>
<td>35 - 40°</td>
<td>5311</td>
</tr>
<tr>
<td>40 - 45°</td>
<td>5893</td>
</tr>
<tr>
<td>45 - 50°</td>
<td>2860</td>
</tr>
<tr>
<td>50 - 54.7°</td>
<td>520</td>
</tr>
</tbody>
</table>

**Angle Ranges from the $\langle 100 \rangle$ Zone**

In this case, latitude areas where calculated from the equator upward and multiplied by a correction factor. The area from the equator to a latitude angle $\theta$ is $A = 2\pi r^2 \sin \theta$.

Each zone must be multiplied by a correction factor since the longitude angle range $\psi$ constantly decreases. The angle between latitude limits at the top and bottom of each zone was measured and the average found. For the $0 - 5^\circ$ angle range, for example, the maximum range in longitude is $45^\circ$, the minimum is $39.8^\circ$. The average is $42.4^\circ$. In the same way a correction factor is found for each zone.
The resulting relative area in 5° increments from the \( \{100\} \) zone are given below:

<table>
<thead>
<tr>
<th>Angle Range</th>
<th>Relative Area</th>
<th>Angle Range</th>
<th>Relative Area</th>
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<td>0 - 5°</td>
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<td>1021</td>
</tr>
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<td>10 - 15°</td>
<td>2725</td>
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<td>366</td>
</tr>
<tr>
<td>15 - 20°</td>
<td>2205</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Since the orientations in one stereographic triangle represent all possible crystal orientations, the relative area in any angle range within a stereographic triangle will be proportional to the relative number of randomly oriented grains within that angle range.
APPENDIX F

Metallographic Specimen Preparation

Macro Etching

The ingot was cut, ground to 600 grit and etched in concentrated Keller’s etch* for 20 – 40 seconds. The surface was then washed with soap and water, dried with alcohol, and re-etched for 1 – 2 seconds.

* Concentrated Keller’s etch
10 parts HF
15 parts HCl
25 parts HNO₃
50 parts H₂O

Micro Etching

Samples were prepared as above to 600 grit and then either:

(a) polished to 1 micron alumina powder, or
(b) electropolished for 10 seconds during a Disa Electropol electropolisher.

Concentrated Keller’s etch was used for etching. The specimen was immersed or swabbed briefly (about 1/2 second), then quickly rinsed in a stream of water and dried with alcohol.
APPENDIX G

Analysis of the Relationship Between Dendrite Trace Structure and Dendrite Orientation

A dendrite plane is shown at the right with a coordinate system defined so that the dendrite plane is parallel to the X axis. Plane (001) is parallel to the chill plane and the Z axis is the heat flow direction. The indices of the dendrite planes are (0kl) where k is a function of θ. θ is the angle between plane (0kl) and plane (010); this is the angle that plane (0kl) lies out of heat flow direction.

When examining a columnar ingot, a plane of cut is made perpendicular to the chill surface. These are cuts along planes (h10), where h can take any value depending on the angle of cut ψ. Dendrite traces are formed when these planes (h10) cut dendrite plane (0kl). It is of interest to find the angle ζ between the dendrite trace and the direction [001] as a function of θ and ψ. The angle ζ is observed on polished sections.

The Miller indices of the dendrite trace \( \mathbf{T} \) are found from the cross product of the dendrite plane \( \mathbf{D} \) and the plane of cut \( \mathbf{C} \). The angle between the heat flow direction [001] and the dendrite trace direction \( \mathbf{T} \) is found by taking the dot product of those two vectors. In mathematical notation:
\[ \overrightarrow{T} = \overrightarrow{D} \times \overrightarrow{C} \]

\[ \overrightarrow{D} = ky + z \]

\[ \overrightarrow{C} = hx + y \]

\[ \overrightarrow{T} = \begin{vmatrix} x & y & z \\ 0 & k & 1 \\ h & 1 & 0 \end{vmatrix} = -x + hy - hz \]

Carrying out the dot product between vector \( \overrightarrow{T} \) and the heat flow direction, \( \overrightarrow{H} = \overrightarrow{z} \),

\[ \cos \theta = \frac{hk}{\sqrt{1 + h^2 + h^2 k^2}} \]

The angle \( \theta \) between plane \((0kl)\) and the heat flow direction is found from

\[ \cos \theta = \frac{h}{\sqrt{1 + h^2}} \]

Angle \( \psi \) is the angle between the X axis \([100]\) and the line of intersection \( \overrightarrow{L} \) of plane \( \overrightarrow{C}(hl0) \) with plane \( xy(001) \).

\[ \overrightarrow{L} = \begin{vmatrix} x & y & z \\ 0 & 0 & 1 \\ h & 1 & 0 \end{vmatrix} = -x + hy, \left[1l0\right] \]

\[ \cos \psi = \frac{1}{h^2 + 1} \]

Table G-1 gives \( k \) versus \( \theta \), and \( h \) versus \( \psi \) with \( k^2 \) and \( h^2 \) added for convenience. Table G-1 shows calculated \( \varphi \) versus \( \psi \) for various values of \( \theta \). This has been plotted and is shown in Figure G-1. It can be seen that at low values of \( \theta \), \( \varphi \) is not very sensitive to \( \psi \) when \( \psi \) is
Figure G-1: Appearance of the trace produced by a longitudinal section through a dendritic plane of a columnar dendrite. $\gamma$ is the observed angle between the trace and the heat flow direction. $\psi$ represents various longitudinal sections, with $\psi = 90^\circ$ defined where the plane of cut is perpendicular to the dendrite plane. $\theta$ is the angle between the dendrite plane and the heat flow direction.

Figure G-2: Width of a dendrite trace produced by cutting a dendrite sheet at various angles. The same graph could be applied to apparent dendrite sheet spacing versus cutting angle.
large. It is obvious in a polished section when the dendrite plane is intersected at a low angle (low $\nu$) because the dendrite trace then appears thicker (as a "band"). This is explained below.

**Apparent Thickness of a Dendrite Sheet**

A dendrite is not a mathematical plane; it has some thickness, $\ell_0$. Therefore, only a cut perpendicular to the plane will reveal its true thickness in a polished section. A dendrite sheet is shown schematically in Figure G-2. $\ell = \ell_0 / \cos \phi$ where $\phi$ is the angle that the plane of cut makes with the dendrite plane. $\ell / \ell_0$ is plotted versus $\phi$ in Figure G-2. The angle between the dendrite sheet and the plane of cut, $\phi$ is found from

$$\cos \phi = \frac{K}{\sqrt{h^2 + 1 + \sqrt{k^2 + 1}}}$$

At $\phi = 90^\circ$, $\phi = 0^\circ$; $\phi$ increases as $\phi$ decreases to an extent which depends on $\theta$. For $\theta$ near $0^\circ$, $\phi - 90^\circ - \ldots$
### TABLE G-1

**k Versus \( \theta \) and h Versus \( \psi \)**

<table>
<thead>
<tr>
<th>( \theta )</th>
<th>( \psi )</th>
<th>( k^2 ) or ( h^2 )</th>
<th>( k ) or ( h )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>90</td>
<td>( \infty )</td>
<td>( \infty )</td>
</tr>
<tr>
<td>5</td>
<td>85</td>
<td>130.4</td>
<td>11.42</td>
</tr>
<tr>
<td>10</td>
<td>80</td>
<td>32.167</td>
<td>5.67</td>
</tr>
<tr>
<td>15</td>
<td>75</td>
<td>13.93</td>
<td>3.73</td>
</tr>
<tr>
<td>20</td>
<td>70</td>
<td>7.548</td>
<td>2.74</td>
</tr>
<tr>
<td>30</td>
<td>60</td>
<td>3.</td>
<td>1.73</td>
</tr>
<tr>
<td>40</td>
<td>50</td>
<td>1.42</td>
<td>1.19</td>
</tr>
<tr>
<td>50</td>
<td>40</td>
<td>0.704</td>
<td>0.84</td>
</tr>
<tr>
<td>60</td>
<td>30</td>
<td>0.333</td>
<td>0.576</td>
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<tr>
<td>70</td>
<td>20</td>
<td>0.133</td>
<td>0.365</td>
</tr>
<tr>
<td>75</td>
<td>15</td>
<td>0.0718</td>
<td>0.268</td>
</tr>
<tr>
<td>80</td>
<td>10</td>
<td>0.031</td>
<td>0.176</td>
</tr>
<tr>
<td>85</td>
<td>5</td>
<td>0.00765</td>
<td>0.0875</td>
</tr>
<tr>
<td>90</td>
<td>0</td>
<td>0.</td>
<td>0.</td>
</tr>
</tbody>
</table>

### TABLE G-2

**\( \gamma \) Versus \( \psi \) for Various \( \theta \) Values**

<table>
<thead>
<tr>
<th>( \theta )</th>
<th>( 0^\circ )</th>
<th>( 5^\circ )</th>
<th>( 10^\circ )</th>
<th>( 20^\circ )</th>
<th>( 30^\circ )</th>
<th>( 45^\circ )</th>
<th>( 90^\circ )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>90°</td>
<td>90°</td>
<td>90°</td>
<td>90°</td>
<td>90°</td>
<td>90°</td>
<td>90°</td>
</tr>
<tr>
<td>5</td>
<td>45°7'</td>
<td>63°09'</td>
<td>76°34'</td>
<td>81°02'</td>
<td>80°6'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>26°43'</td>
<td>45°28'</td>
<td>64°36'</td>
<td>75°20'</td>
<td>80°9'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>14°18'</td>
<td>27°10'</td>
<td>46°46'</td>
<td>59°20'</td>
<td>71°6'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>9°56'</td>
<td>19°17'</td>
<td>36°11'</td>
<td>49°12'</td>
<td>63°31'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>7°44'</td>
<td>14°58'</td>
<td>29°26'</td>
<td>42°2'</td>
<td>57°19'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>50</td>
<td>6°34'</td>
<td>12°58'</td>
<td>25°42'</td>
<td>37°1'</td>
<td>52°29'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>60</td>
<td>5°45'</td>
<td>11°30'</td>
<td>22°45'</td>
<td>33°37'</td>
<td>49°5'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>5°25'</td>
<td>10°41'</td>
<td>21°09'</td>
<td>31°41'</td>
<td>47°0'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>80</td>
<td>5°14'</td>
<td>10°12'</td>
<td>20°15'</td>
<td>30°28'</td>
<td>45°25'</td>
<td>90°</td>
<td></td>
</tr>
<tr>
<td>90</td>
<td>5°</td>
<td>10°</td>
<td>20°</td>
<td>30°</td>
<td>45°</td>
<td>90°</td>
<td></td>
</tr>
</tbody>
</table>
APPENDIX H

Measurements of Dendrite Growth Form Spacing in Columnar Dendrites

The structure of columnar dendrites has been described in Chapter III. Columnar dendrites become aligned with a $[100]$ direction nearly perpendicular to the chill. (For simplicity of discussion the $[100]$ direction will be assumed to be exactly perpendicular to the chill.) The $[100]$ direction is at the intersection of two $<100>$ planes (the (010) plane and (001) plane). Crystallographically, therefore, two $<100>$ planes are perpendicular to the chill and one (the (100) plane) parallel to the chill.

The dendrite growth forms are parallel to $<100>$ planes in the crystal. Primary dendrite sheets are perpendicular to the chill, and lie in (010) and (001) planes. Secondary sheets, branching off the primary sheets, lie in (100) planes parallel to the chill. Tertiary sheets lie in the same planes as primary sheets; and are usually attached to secondary sheets. Sometimes "tertiary sheets" may be attached to primary sheets. Attachment implies branching of one growth form from the other, and the connotation of the word "tertiary" is a branching from "secondary" rather than from primary. However, the classification used here is based on orientation rather than attachment, because this is a more convenient basis.

Columnar ingots are usually examined by sectioning transversely (parallel to the chill surface) and longitudinally (perpendicular to the chill surface). Considering the geometry of primary, secondary and tertiary dendrite sheets, it is clear how the growth form spacings should
be determined. Primary spacings are determined on a transverse section; in this section the spacing between traces in dendrite "crosses" or dendrite "lines" is an accurate measure of the spacing between dendrite sheets (see Figure G-2). A longitudinal section will give a variety of results depending on how the cut is taken. For the same reason, tertiary spacing should also be measured on a transverse section. On the other hand, secondary spacing must be measured on a longitudinal section, which lies perpendicular to secondary sheets. A transverse section can never reveal secondary spacing because the plane of cut in this case is parallel to the dendrite plane. See Figure H-1 for a graphical picture of dendrite growth form spacing.
Figure H-1: Relationship between primary, secondary and tertiary dendrite growth form spacings in longitudinal and transverse sections of a columnar ingot. See also Figure G-1 and G-2. ψ is defined in Appendix G. "P" is the primary sheet spacing. In a real ingot average values of primary, secondary and tertiary spacings are determined.
APPENDIX J

Solidification Times of Columnar Ingots 1, 2 and 3 and Dendrite Growth Form Spacing in Columnar Ingot 3

TABLE J-1

Cooling Times from 1200°F to 1150°F (Seconds)

<table>
<thead>
<tr>
<th>Distance from Chill</th>
<th>Ingot 1</th>
<th>Ingot 2</th>
<th>Ingot 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chill</td>
<td>8</td>
<td>1740</td>
<td>370</td>
</tr>
<tr>
<td>1&quot;</td>
<td>37</td>
<td>2520</td>
<td>650</td>
</tr>
<tr>
<td>2&quot;</td>
<td>62</td>
<td>3180</td>
<td>-</td>
</tr>
<tr>
<td>3&quot;</td>
<td>99</td>
<td>3480</td>
<td>990</td>
</tr>
<tr>
<td>5&quot;</td>
<td>190</td>
<td>4500</td>
<td>1170</td>
</tr>
<tr>
<td>7&quot;</td>
<td>292</td>
<td>4260</td>
<td>1150</td>
</tr>
<tr>
<td>10&quot;</td>
<td>-</td>
<td>4800</td>
<td>1710</td>
</tr>
</tbody>
</table>

TABLE J-2

Dendrite Growth Form Spacings in Columnar Ingot 3 (Microns)

<table>
<thead>
<tr>
<th>Distance from Chill</th>
<th>Primary</th>
<th>Secondary</th>
<th>Tertiary</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chill</td>
<td>-</td>
<td>113</td>
<td>145</td>
</tr>
<tr>
<td>1&quot;</td>
<td>925</td>
<td>151</td>
<td>150</td>
</tr>
<tr>
<td>3&quot;</td>
<td>925</td>
<td>185</td>
<td>150</td>
</tr>
<tr>
<td>5&quot;</td>
<td>925</td>
<td>167</td>
<td>150</td>
</tr>
<tr>
<td>7&quot;</td>
<td>1000</td>
<td>208</td>
<td>150</td>
</tr>
<tr>
<td>10&quot;</td>
<td>1100</td>
<td>275</td>
<td>167</td>
</tr>
</tbody>
</table>