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### MACROSEGREGATION CHARACTERISTICS OF COMMERCIAL SIZE ALUMINUM ALLOY INGOT CAST BY THE DIRECT CHILL METHOD

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#### Abstract

A metallurgical investigation of the macrosegregation occurring in commercial size direct chill cast Al-Zn-Mg-Cu alloy ingot is described. The extent of the chemical homogeneity from the surface to the center of the rectangular ingot produced by the bi-level transfer method is compared to ingot produced by the level pour method. The relative severity of macrosegregation for each major alloying element is discussed. Existing metallurgical theories of macrosegregation including interdendritic liquid flow and convection in the liquid pool are used to explain the observed variation in chemical composition. An analysis is carried out to qualitatively illustrate the importance of the volume fraction of the detached dendrites and the partition coefficient of each major alloying element contributing to the level of negative macrosegregation at the centerline of the ingot.

#### Introduction

The macrosegregation occurring in ingot during solidification has a significant impact both on the composition and structural homogeneities of the finished product. Several studies on this solidification phenomenon have been carried out to analyze the mechanisms responsible for the macrosegregation and quantitatively predict the level of macrosegregation. In 1967, Flemings et al.<sup>1-3</sup> developed expressions for macrosegregation due to the movement of liquid within the mushy zone during solidification. The driving force for the interdendritic flow includes thermal contractions, solidification shrinkage and the density differences in the interdendritic liquid. This work has greatly advanced our understanding of macrosegregation phenomena. Subsequently, in a work on axisymmetric laboratory ingot, Ridder, Kou and Mehrabian<sup>4</sup> coupled bulk flow in the metal pool to interdendritic flow during solidification. Results of this work indicate that natural convection in the liquid pool had little effect on the interdendritic fluid flow of a bench scale ingot.

Although the liquid movement within the mushy region appears to be by far the most important and general cause of segregation, floating and settling of precipitated phases early in solidification can become an important cause of macrosegregation in some situations such as casting of commercial size ingots where the natural convection in the liquid pool can be very strong. Recently, Yu and Granger<sup>5,6</sup> analyzed the macrosegregation occurring in commercial size direct chill (DC) aluminum ingot cast by the bi-level transfer method and concluded that the natural convective flow in the molten metal pool plays an important role in accounting for the negative macrosegregation observed at the center of the large ingot. Their argument was supported by the observation of coarse dendrite cells within the fine dendrite cells at the center of the ingot. The coarse cells were thought to originate from dendrites formed early in the solidification process which detached and were carried by the strong convection current into the pool and then grew isothermally within the thermal boundary layer close to the liquidus and finally become entrapped in the solidifying ingot at the bottom of the pool.

This paper describes recent results on the composition and microstructure analysis from the surface to the center of 16 in. x 50 in. Al-Zn-Mg-Cu alloy rectangular ingot cast by both the bi-level transfer and level pour method. In general, ingot produced by the bi-level transfer method would be expected to have a stronger convective current flow in the liquid pool than ingot produced by the level pour method. The difference observed between the two ingot types is discussed and explanations for the unique observations in this study are presented.

#### **Experiments**

Figure 1 is a schematic drawing of the direct chill casting process using the bi-level transfer and level pour method, respectively. In the bi-level method, molten metal is transferred from the pouring trough (high level) to the mold (low level). In the level pour method, the metal is transferred from the trough to the mold without a change in elevation. An insulation basin prevents solidification above the top of the mold.

The locations of samples taken from each ingot for composition and microstructure analysis are shown in Figure 2. In general, two sets of samples were taken across the ingot thickness at the (width)/2 location. One set of samples was remelted and chill cast prior to determining the chemical composition on the quantometer. The other set of samples was polished and examined using optical metallography.

#### **Results and Discussion**

#### 1. Macrosegregation Characteristics and Microstructure

The typical macrosegregation profiles of 16 in. x 50 in. Al-Zn-Mg-Cu direct chill ingot cast by the bi-level transfer and level pour methods are shown in Figure 3, respectively. The Y axis represents the deviation of each major element from the melt chemistry. These profiles exhibit a similar pattern regardless of casting method or element type.

The region adjacent to the ingot surface is highly enriched with solute. The microstructure at this region is comprised of two distinct morphologies. One is an equiaxed dendrite type (designated as region A in Figure 4) and the other type is fine dendrites surrounded by a high fraction of eutectic (designated as region B in Figure 4). Region B is a result of penetration of highly enriched liquid through the coherent ingot shell following shrinkage of the ingot due to solidification and resultant loss of cooling. On the other hand, region A is a result of liquid flow to feed shrinkage (i.e., inverse segregation). Nevertheless, both

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#### The Locations of Samples Taken from Each Ingot for Composition and Microstructure Analysis Figure 2

types of structures contribute to the sharp increase of solute content for this region.

Away from the subsurface solute enriched region, a region with negative segregation is observed. The cause of this negative segregation is believed to be related to the sudden increase in the rate of heat extraction when the ingot shell enters the direct chill zone at the bottom of the mold. Note that there is an air gap between the ingot shell and the mold prior to entering the direct



Distance from ingot surface across ingot thickness (inch)

#### Macrosegregation Profiles of 16"×50" Al-Zn-Mg-Cu Ingot Cast by the Bi-level and Level Pour Methods. The y Axis Represents the Deviation of Each Major Alloy Elements from the Melt Chemistry Figure 3

chill water cooling zone. The sudden increase in the rate of heat extraction is indicated by the decrease in cell size at this region. The measured cell size versus the distance from the ingot surface for the ingot cast by the level pour method is shown in Figure 5. The change in cell size for the region mentioned above is marked with an arrow in the figure. Flemings et al.<sup>3</sup> reported previously that a sudden increase in the rate of heat extraction led to a marked negative segregation in a unidirection solidified laboratory scale ingot. According to their analysis, a sudden increase in the rate of heat extraction of the width of the liquid-solid region and thereby leads to the negative segregation.

As shown in Figure 5, except for the region at the distance less than one inch from the ingot surface the cell size increases with distance from the ingot surface. As-cast microstructures for the regions corresponding to the T/16, 5T/16 and T/2 (i.e., center of the ingot) locations, respectively, are shown in Figure 6 for comparison. Note that the microstructure at or near the center of the ingot comprises a mixture of coarse cells and fine dendrites. The average cell size of coarse cells is  $80 \ \mu m$  while the average cell size of fine dendrites is  $40 \ \mu m$ . The origin of fine dendrite and coarse cells will be discussed later.

As shown in Figure 3, ingot cast by the bi-level and level pour methods exhibits a high level of negative segregation at the centerline of the ingot. The ingot cast by the level pour method has a 15% improvement in the maximum negative deviation (i.e.,  $(C_n - C_m)/C_n$ , where  $C_n$  is the nominal composition, and  $C_m$  is the maximum negative segregation) over the ingot cast by the bilevel transfer method. In addition, the extent of the centerline negative macrosegregation is only about 20% of the total ingot thickness for ingot cast by the level pour method while the extent of centerline negative segregation for the ingot cast by the bi-level transfer method is over 30% of the total ingot thickness. As a result of using a peripheral feed arrangement for the ingot cast by the level pour method, the flow of the molten metal entering the basin area was more uniform than the ingot cast by the bi-level transfer method. As a result, the liquid pool in the ingot cast by the level pour method would be expected to have a less severe convective current flow than the ingot cast by the bi-level transfer method. Therefore, current results indicate that the degree and extent of the centerline negative segregation are strongly affected by the convective current flow in the liquid pool. This finding confirms the hypothesis proposed previously by Yu and Granger<sup>5</sup> that the natural convective current flow in the liquid pool accounts for the negative macrosegregation at the centerline of the ingot. However, judging from the cell size--distance from chill relationship shown in Figure 5--the fine dendrites, as shown in Figure 6C and in Figure 7 at a smaller magnification, should be those dendrites detached early in the solidification that settle at or near the center of the ingot. Since fine dendrites formed early in the solidification are solute poor, they are responsible for the negative segregation at these regions. This explanation of the origin of the fine dendrites does not agree with the conclusion previously reached by Yu and Granger, namely, that the coarse cells originated from the detached dendrites. In addition to the argument provided above, our conclusion of fine dendrites at the center of the ingot is further supported by an another observation: the fine dendrites at the centerline of the ingot cast by the level pour method are about 5% less by volume than the ingot cast by the bi-level transfer method.

#### 2. The Analysis

To illustrate the importance of volume fraction of detached dendrites and the partition coefficient of each major alloying element contributing to the level of negative macrosegregation at the centerline of the ingot, the following assumptions are made to estimate the average composition for the regions at the centerline of the ingot:

- 1. The structure at the center of the ingot comprises a mixture of fine dendrites and coarse cells. The fine dendrites originated from the start of the solidification and the coarse cells grew in situ.
- 2. The fine dendrites are detached by the convective flow before they are fully developed.
- 3. There is no growth of the detached dendrites during settling to the center of the ingot.
- 4. The coarse cells grew in situ from a liquid of initial composition.
- 5. The detached dendrites are cylindrical.



The As-Cast Microstructure for the Region Adjacent to the Ingot Surface. The Region Designated as A is Equaixed Dendritic Cells and the Region Designated as B is Fine Dendrites Surrounding by a High Fraction of Eutectic Figure 4



The Measured Cell Size versus the Distance from Ingot Surface to the Center of a  $16"\times 50"$ Level Pour Ingot. The Change in Cell Size as Indicated by an Arrow is Related to the Sudden Change Increase in the Rate of Heat Extraction when the Ingot Shell Enters the Direct Chill Zone at the Bottom of the Mold. Figure 5

Now, the average composition of cellular dendrites ( $\overline{C}_{isd}$ ) solidified in situ at the centerline of the ingot can be calculated by the following expression :

$$\bar{C}_{isd} = \frac{\rho_s \int_0^{1-f_E} C_s df_s + \rho_{sE} f_E C_E}{\rho_s (1-f_E) + \rho_{sE} f_E}$$
(1)

where  $\rho_s = \text{solid density}, g/cc$ 

 $C_s$  = interface composition, weight fraction

- $f_s =$  volume fraction of solid
- $f_E$  = volume fraction of eutectic

 $\rho_{sE}$  = density of eutectic, g/cc

 $<sup>\</sup>bar{C_E}$  = composition of eutectic, weight fraction



As-Cast Microstructures for the Regions Corresponding to (a) (1/16)T (b) (5/16)T and (c) T/2 Locations. (T Represents 16 Inch Thick Ingot) Figure 6

If the Scheil type solidification is followed, the interface composition of solid ( $C_s$ ) at each fractional distance can be expressed as:

$$C_{s} = \kappa C_{0} \left(1 - f_{s}\right)^{\kappa - 1}$$
(2)

where  $\kappa = equilibrium partition ratio C_o = initial composition, weight fraction$ 

Figure 8 shows a calculated solute redistribution profile across a cell or secondary arm using the above equation for an Al-2.5% Cu alloy. For a fully developed dendrite arm or cell ,the average composition of the solid can be obtained by integrating the area under the curve from  $f_s = 0$  to  $f_s = 1$ -  $f_E$  and dividing the integral by 1 -  $f_E$ . Now, if a dendrite arm is detached at the fraction solid  $f_s$  in the early stages of solidification before it is fully developed, then the average composition of the detached dendrite ( $\bar{C}_{dd}$ ) can be calculated by:

$$\bar{C}_{dd} = \frac{\int_{0}^{f_{s}} C_{s} df_{s}}{f_{s}}$$

$$= \frac{\int_{0}^{f_{s}} k C_{0} (1 - f_{s})^{k - 1} df_{s}}{f_{s}}$$

$$= \frac{C_{0} \left[1 - (1 - f_{s})^{k}\right]}{f_{s}}$$
(3)

Assume that the structure at the centerline of the ingot comprises  $g_{dd}$  fine dendrites which are detached in the early stages of solidification and settled to the ingot center and  $(1 - g_{dd})$  coarse cells which solidified in situ by volume, then the average composition of solid ( $\overline{C}_s$ ) at this region will equal to

$$\overline{C}_{s} = \overline{C}_{dd} \cdot g_{dd} + \overline{C}_{isd} \cdot (1 - g_{dd})$$
(4)

Since  $\bar{C}_{isd}$  is equal to the initial composition of the liquid  $C_o$  is assumed, then Equation 4 becomes

$$\overline{C}_{s} = \frac{C_{o} \left[ 1 - (1 - f_{s})^{k} \right] \cdot g_{dd}}{f_{s}} + C_{o} \cdot (1 - g_{dd})$$
$$= C_{o} \left[ 1 + \frac{(1 - f_{s}) - (1 - f_{s})^{\kappa}}{f_{s}} \cdot g_{dd} \right]$$
(5)

#### 2.1 The Effect of Volume Fraction of Detached Dendrites

Figure 9 shows the calculated average composition and negative deviation from the nominal composition at the centerline of the ingot as a function of volume fraction of detached dendrites using Equation 5. The calculations were made assuming that the dendrites solidified at the start of the solidification were broken up at three different fractions of a fully developed cell or secondary dendrite arm. The results indicate that the level of the negative segregation is strongly affected by the volume fraction of the detached cellular dendrites present at the centerline of the ingot for a given fraction of a fully developed cell or secondary dendrite arm. The measured volume fraction of detached dendrites at the centerline of the ingot is about 15% for ingot cast by the level pour method and 20% for ingot cast by the bi-level transfer method. These yield deviations of 9.5% and 10.6% from the nominal composition (2.5% Cu) approximately using the curve for  $f_s = 0.90$  in Figure 9 or Equation 5. These estimated values are slightly over the measured values shown in Table 1. This small discrepancy may be associated with some flow of enriched liquid to feed shrinkage and the possible continuous growth of the detached dendrites that settled and were trapped in the mushy zone at the center of the ingot.

#### 2.2 The Effect of Partition Coefficient

In Figure 3 and Table 1, it appears that copper exhibits the largest negative deviation from the nominal composition for the regions at the centerline of the ingot followed by magnesium and zinc. It is believed that the degree of the deviation is directly related to the partition coefficient. This is illustrated in Figure 10 which shows the calculated average compositions and the deviations from the initial composition of 2.5 wt. % as a function of volume fraction of the detached dendrites for different partition coefficients. The

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#### A Mixture of Coarse Cells and Fine Dendrites at the Center of the Ingot Figure 7

calculations were made assuming that the partition coefficient is a constant. The partition coefficients of Cu, Mg and Zn in aluminum binary systems are 0.171, 0.285 and 0.45 (indicated by arrow signs in Figure 10), respectively. If it is assumed that the partition coefficients of Cu, Mg and Zn in the Al-Zn-Mg-Cu system are similar to that in binary systems, the calculation results indicate that the average composition at the center of the ingot decreases as the value of the partition coefficient among all the major alloying elements, copper would be expected to have the largest negative deviation.

#### **Conclusions**

- 1. The measured macrosegregation profiles of 16 in. x 50 in. Al-Zn-Mg-Cu alloy ingots exhibit similar patterns regardless of casting method or alloying element type. Each profile exhibits a solute enriched zone at the subsurface, a negative segregation at 1 in. from the ingot surface and a high degree of negative segregation at the centerline of the ingot.
- 2. The ingot cast by the level pour method has approximately a 15% improvement in the negative segregation (particularly for Cu) at the centerline of the ingot over the ingot cast by the bi-level transfer method due to the more uniform distribution of molten metal entering the basin area during casting.
- 3. The microstructure at the center of the ingot consists of a mixture of coarse cells and fine dendrites. The fine dendrites originated in the early stages of the solidification and detached as a result of the convective current flow in the metal pool. They account for the centerline negative segregation.
- 4. A simplified analysis is proposed. The analysis indicates that the level of centerline negative segregation is strongly affected by the volume fraction of detached cellular dendrites and the partition coefficient of each major alloying element. The predicted values in this work agree with the experimental results.

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# Table 1Deviation from Nominal Composition<br/>at the Center of the Ingot

	Bilevel transfer (%)	Level pour (%)
Cu	-10.3	-8.5
Mg	-7.8	-5.4
Zn	-5.5	-3.0



Calculated Solute Redistribution Across a Secondary Arm Using the Scheil Equation for AI-2.5 wt% Cu Alloy Figure 8



Calculated Average Composition and Negative Deviation from the Nominal Composition at the Centerline of the Ingot as a Function of Volume Fraction of Detached Dendrites Figure 9



Calculated Average Composition and Negative Deviation from the Nominal Composition at the Centerline of the Ingot as a Function of Partition Coefficent Figure 10