Mechanism for Complex Morphology Due to Mechanical Vibration

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The feasibility of reducing macrosegregation via control of the solidified structure was investigated. Factors such as the size and complexity of equiaxed grains, which may influence this macrosegregation, were determined. In the first stage of the project, the influence of mechanical vibration on the complexity of the solidified structure was determined. Al-10 wt.%Cu alloys were used as model alloys. The size of the equiaxed grains varied with the amount of modifier. In addition, the complexity of the morphology of primary aluminum was characterized in terms of the fractal dimension, which was measured by using the box-counting method. The results revealed that the fractal dimension increased with increasing frequency of the vibration. Moreover, the diffusion layer in front of the growing interface may be washed away, owing to this vibration. The consequent decrease in thickness of this layer resulted in an increase in the degree of constitutional undercooling. Therefore, the break-down may occur, and hence complexity of the solid/liquid interface may increase. The results also revealed that the size and complexity of the equiaxed grains may be independently controlled to some extent.

KEY WORDS: equiaxed grains; vibration; permeability; fractal dimension; Al–Cu alloy; solidification; casting; macrosegregation; secondary dendrite arm spacing; modifier.

1. Introduction

Macroscopic segregation in steel semi-final products may be governed by liquid flow during the final stage of solidification; this flow results from additional forces, such as bulging and volume contraction during solidification.1) Centerline segregation in continuously cast slabs may be reduced by the soft reduction imparted by rolls in the strand; this reduction compensates for the volume change resulting from solidification.2,3) Analysis of the resistance to fluid flow in regions with high solid fraction is essential for preventing centerline segregation. This resistance, i.e., the “permeability” has been extensively investigated. For example, using the definition of the parameter, Piwonka and Flemings4) attempted direct measurements of the permeability in a metallic system. Murakami et al.5,6) measured the permeability of an organic alloy system, which consisted of water as a liquid phase. They varied the solidified structure and changed the direction of fluid flow. In addition, they found that the permeability varies with the direction of fluid flow, i.e., the permeability along the primary dendrites is lower than that perpendicular to the primary dendrites.

Natsume et al.7) created three-dimensional (3D) dendrites based on the calculation results of phase field simulations, and determined the permeability via fluid-flow calculations of the residual liquid phase in the interdendritic region. In that work, flow directions (parallel or normal to the columnar dendrite) were taken into consideration. The simulation results corresponded closely to the experimental results of Murakami et al.,5,6) although the calculations considered low solid fractions only. This indicated that the permeability can be estimated from fluid-flow calculations in porous media. Furthermore, fluid flow in the interdendritic region was expressed through a unique relation that was valid over the wide range of solid fraction. They also showed that the permeability varies with the solid fraction and the size of the features in the solidified structure. For example, for the same solid fraction, the permeability increases with decreasing primary dendrite arm spacing. This suggests that the permeability increases with decreasing size of the equiaxed grains; the decrease results from the narrow and intricate path through which the residual liquid moves even at constant volume fraction of the primary phase. For the same reason, the permeability may also increase with increasing complexity in the shape of the equiaxed grains.

Therefore, the aim of our research project is to determine the feasibility of controlling macroscopic segregation via control of the size and complexity of the equiaxed grains. The first step of the project includes a fundamental study in which the possibility of independently controlling the size and complexity is explored, using Al–Cu alloy as a model alloy. In addition, the effect of mechanical vibration on the morphology of the equiaxed grains was investigated.

2. Experimental Procedure

An Al–Cu alloy was used as a model alloy in this study. This alloy was selected for the following reasons: the melt-
ing temperature of the alloy is around 620°C and therefore the alloy can be easily handled. The macroscopic solidified structure can be changed from columnar to equiaxed grains by using a modifier, which contains TiB
2 and/or TiAl
3 and therefore promotes heterogeneous nucleation of primary aluminum. In addition this structure is easily revealed via backscattered electron images (BSE images), even without etching. Al-10 wt.%Cu (200 g) alloy was prepared from the raw metals; both metals had a purity of 4N. The alloy was melted under Ar atmosphere. Immediately prior to casting, the modifier was added to the molten aluminum alloy, in amounts ranging from 0.005 wt.%–0.2 wt.%. The alloy was cast at a fixed temperature of 800°C and poured into a 50 mm (outer diameter) × 40 mm (inner diameter) × 100 mm (height) alumina mold (i.e., crucible). Half of the molten metal was poured into an alumina mold, which was tightly affixed to the vibrator. The other half was poured into an alumina mold, which was placed on the floor. Mechanical vibration was applied to the mold, immediately after casting. This vibration was applied by an apparatus that was originally used for endurance testing of electrical devices, and has the ability to apply vertical sinusoidal mechanical vibration. The amplitude (A) and frequency (f) of the vibration are independently selected and controlled by a PC. The A and f used in this study are listed in Table 1.

Longitudinal 30 mm × 40 mm cross-sections of the specimen were prepared for metallographic characterization. The specimens were polished with various grades of emery paper of up to #4000 and subsequently examined by using a field-emission scanning electron microscope (FE-SEM).

<table>
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<th>Vibration condition used in this study.</th>
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<tr>
<td>Frequency, f (Hz)</td>
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<td>Amplitude, A (mm)</td>
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Large equiaxed grains were selected and the diameter of these grains were measured. These grains were approximately spherical and hence, a cut plane passing through the center of a grain should correspond to the maximum size.

In this study, the complexity of the primary aluminum was characterized in terms of the fractal dimension. Fractal dimension was determined by using a box-counting method, where the perimeter of the equiaxed grain was limited to a width of 1 pixel. The determined value exceeds the real value at widths of > 1 pixel. Therefore, the width was carefully adjusted in order to minimize the error of the measurement and at least five measurements were performed for each observation plane.

The solidified structure was correlated with the cooling condition by determining the thermal histories of the casting samples with and without vibration. Here, 0.1 mm φ chromel-alumel thermocouples were used and the hot junctions were thinly coated by alumina cement, in order to keep sensitivity. Each thermocouple was firmly affixed by a special screw to prevent shifting during mechanical vibration and it was placed 10 mm away from the wall of the mold. In addition, the experimental data was digitally recorded in 5 ms intervals.

3. Experimental Results

3.1. Solidified Structure

Figure 1 shows typical examples of the solidified structure obtained at a modifier content of 0.005 wt.%. As indicated in the figure, four conditions (three with vibration and one without) were considered. These BSE images show the central region of the specimen. Gray or dark crystals and white regions correspond to primary aluminum and the eutectic structure, respectively. High-magnification views of the latter region reveal a lamellar structure that consists of the white Al-Cu phase and the gray aluminum phase. This lamellar structure was only visible at high magnification.

Fig. 1. BSE images of solidified structure in Al-10 wt.%Cu alloy when the amount of modifier was 0.005 wt.%. a) without vibration, b) f = 50 Hz, A = 0.5 mm, c) f = 100 Hz, A = 0.1 mm and d) f = 500 Hz, A = 0.05 mm.
Primary aluminum exhibited a coarse dendritic structure, which had a high aspect ratio, and was therefore judged non-equiaxed grains. This type of solidified structure is typically referred to as 'broken columnar dendrite'.

Figure 2 shows the solidified structures formed in the central region of the specimen consisting of 0.05 wt.% of the modifier. A comparison of Figs. 1 and 2 reveals that the primary grains in the 0.05 wt.%-added specimen are significantly smaller than those in the 0.005 wt.%-added specimen. This refinement is attributed to the effect of the modifier. Furthermore, the aspect ratio of the primary grains is close to unity and these grains are therefore deemed isotropic equiaxed grains. Secondary or higher-order dendrite arms are also formed. More importantly, the solidified structure seems to be refined with increasing frequency of vibration.

The solidified structure of the specimens consisting of 0.2 wt.% of modifier is shown in Fig. 3. As in the case of Figs. 1 and 2, four conditions (three with vibration and one without) were considered. The primary crystals were granular and did not exhibit a typical dendritic structure. In fact, these equiaxed grains were the smallest obtained in this study. The size of the primary grains remained approximately constant regardless of the vibration condition, whereas the dendrite arms were refined with increasing frequency of vibration.

Fig. 2. BSE images of solidified structure in Al-10 wt.%Cu alloy when the amount of modifier was 0.05 wt.%. a) without vibration, b) $f=50$ Hz, $A=0.5$ mm, c) $f=100$ Hz, $A=0.1$ mm and d) $f=500$ Hz, $A=0.05$ mm.

Fig. 3. BSE images of solidified structure in Al-10 wt.%Cu alloy when the amount of modifier was 0.2 wt.%. a) without vibration, b) $f=50$ Hz, $A=0.5$ mm, c) $f=100$ Hz, $A=0.1$ mm and d) $f=500$ Hz, $A=0.05$ mm.
3.2. Characterizations of the Solidified Structures

Figure 4 shows a magnified view of the solidified structure obtained at a modifier content, \( f \) and \( A \) of 0.05 wt.\%, 500 Hz and 0.05 mm, respectively. An equiaxed grain was selected as indicated in the figure. This grain has a round and large envelop and the corresponding circle-equivalent diameter of 162 \( \mu \)m was calculated by using a software. The periphery of the equiaxed grain was then extracted and the width of the line was adjusted to one pixel, as shown in Fig. 4. Subsequently, the fractal dimension (i.e., 1.332) of the grain was measured via the box-counting method.

BSE images of the solidified structure were obtained, at a suitable magnification from twelve predetermined areas including the central and near-surface regions. At least five grains were selected from each image and the corresponding equivalent diameters and fractal dimensions were measured, in accordance with the aforementioned procedure.

The grains associated with the previously mentioned ‘broken columnar dendrites’, which formed at a modifier content of 0.005 wt.\%, were larger than the viewing area. Therefore, the grain diameter and fractal dimension could not be measured.

Figure 5 shows the average diameter of the primary phase as a function of the frequency of vibration, for a modifier content of 0.05 wt.\%. As the figure shows, a constant grain diameter is obtained, irrespective of the frequency. This indicates that the diameter is determined by the amount of modifier, and remains constant with the vibration after casting. The results of fractal dimension measurements of the

equiaxed grains are shown in Fig. 6. The fractal dimension initially decreased at low frequency, and increased thereafter with increasing frequency. In the case of low frequency, the method used to characterize the morphology of the primary phase will be discussed in section 4.3.

Figure 7 shows the average diameter of the equiaxed grains as a function of the frequency of vibration at a

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**Fig. 4.** Examples of characterization for equiaxed grain: equivalent diameter \( d \) and fractal dimension \( D_f \).

**Fig. 5.** Relationship between frequency of vibration and equivalent diameter of equiaxed grain when the amount of modifier was 0.05 wt.\%.

**Fig. 6.** Relationship between frequency of vibration and fractal dimension of equiaxed grain when the amount of modifier was 0.05 wt.\%.

**Fig. 7.** Relationship between frequency of vibration and equivalent diameter of equiaxed grain when the amount of modifier was 0.2 wt.\%.

**Fig. 8.** Relationship between frequency of vibration and fractal dimension of equiaxed grain when the amount of modifier was 0.2 wt.\%.
modifier content of 0.2 wt.%. As in the case of 0.05 wt.% of modifier, the grain diameter remained almost constant with changing frequency. This indicates that the vibration after casting has no effect on the size of the equiaxed grains. The corresponding fractal dimension of each equiaxed grains is shown in Fig. 8. In spite of the scatter in the experimental data, the figure shows that the fractal dimension increases monotonically with increasing frequency.

The solidified structure was also characterized by measuring the secondary dendrite arm spacing ($\lambda_2$). A globular structure was obtained at a modifier content of 0.2 wt.% and, therefore, the secondary dendrite arm could not be defined. As such, Fig. 9 shows the dependence of $\lambda_2$ on the frequency at modifier contents of 0.005 and 0.05 wt.%. $\lambda_2$ was almost constant at 0.005 wt.% of modifier independent of the frequency. On the other hand, $\lambda_2$ decreased with increasing frequency at 0.05 wt.% of modifier.

### 3.3. Thermal History

An example of the thermal history, with and without vibration is shown in Fig. 10. In this case, the amount of modifier, $f$ and $A$ were 0.2 wt.%, 500 Hz, and 0.05 mm, respectively. In order to easily compare the cooling behavior, these results were adjusted so that the ‘casting’ time was the same. Initially, the temperature increased abruptly, owing to the pouring of the hot molten alloy and decreased monotonically thereafter. The liquidus and eutectic temperatures are shown in the figure; small thermal arrests occur at these temperatures. Primary solidification of aluminum begins at the initial point of decrease from the liquidus temperature and ends when the eutectic temperature is reached. The cooling rates from the liquidus temperature to the eutectic temperature, which would determine the secondary dendrite arm spacing, were calculated. Rates of 5.2°C/s and 5.1°C/s, corresponding to solidification with and without vibration, respectively, were obtained; the difference (i.e., 0.1°C/s) lies within the error of the measurement and hence, these rates are deemed equal. Furthermore, as the arrow in Fig. 10 indicates, the end-point of eutectic solidification was the same, irrespective of the vibration condition. The mechanical vibration had, therefore, no effect on the cooling condition.

![Fig. 9. Relationship between frequency of vibration and secondary dendrite arm spacing. 1) When the amount of modifier was 0.005 wt.%. 2) When the amount of modifier was 0.05 wt.%](image)

![Fig. 10. Thermal histories of molten Al-10 wt.%Cu alloy cast in the alumina mold with and without mechanical vibration when the amount of modifier was 0.2 wt.%. The position of thermocouple was 10 mm from the alumina wall.](image)

![Fig. 11. Relationship between the amount of modifier and equivalent diameter of equiaxed grain.](image)

### 4. Discussions

#### 4.1. Factors Affecting the Equiaxed Grain Size

As Figs. 5 and 7 show, the diameter of the equiaxed grains is constant and independent of the vibration condition. This indicates that nucleation of the aluminum phase was not enhanced under the present vibration conditions. Figure 11 shows the average diameter of the equiaxed grains, as a function of the amount of modifier. The modi-
fier contains TiB$_2$ and/or TiAl$_3$ particles, which may act as heterogeneous nucleation sites for primary aluminum. Satou et al.$^{9}$ formulated a relationship between the amount of modifier ($w$) and the average diameter of an equiaxed grain ($d$); this formulation is based on the assumption that the probability of nucleation is independent of the number density of nucleation sites.

$$d \propto w^{-\frac{1}{3}}$$ .......................... (1)

The power of the present empirical equation is $-0.38$, which corresponds closely to the result of the aforementioned model. This confirms that the diameter of the equiaxed grains is determined by the amount of modifier and the mechanical vibration has no effect on the nucleation of the primary aluminum phase.

4.2. Factors Affecting the Complexity of the Equiaxed Grains

During growth of the primary phase, the solute element is continuously rejected at the solid/liquid interface, if the partition ratio ($k$) is less than unity. This results in a piled-up of solute at the interface as shown schematically in Fig. 12. Therefore, this causes constitutional undercooling as well as the break-down at the solid/liquid interface.$^{11}$ These occurrences may lead to the formation of a dendritic structure. Mechanical vibration of the semi-solid may result in relative motion of the liquid phase with respect to the solid, owing to the difference in the density of these phases.$^{12}$ In fact, at its melting point, the density of solid aluminum is approximately 6% higher than that of liquid aluminum.$^{13}$ As shown in Fig. 13, the piled-up solute may be washed away by the motion-induced flow at the solid/liquid interface.

The solutal fields around the solid particle shown in Figs. 12 and 13 indicate that the degree of constitutional undercooling in a vibrated liquid may be larger than that in a stagnant liquid. Therefore, as schematically illustrated in Fig. 14, the solid/liquid interface may become complex when mechanical vibration is applied. Liquid flow (based on this vibration), and hence the complexity of the equiaxed grain, may increase with the frequency of vibration. Accordingly, the fractal dimension increased with increasing frequency of vibration, as shown in Figs. 6 and 8.

In contrast, the secondary dendrite arm spacing ($\lambda_2$) decreased with increasing frequency of vibration. At a modifier content of 0.005 wt.%, however, $\lambda_2$ remained constant, even when mechanical vibration was applied (Fig. 9 1)). In this case, the relative motion of solid and liquid phases did not occur owing to the entangled broken columnar grains so that there was no change in morphology.

Figure 9 2) showed that the secondary dendrite arm spacing decreased with mechanical vibration. This indicates that the cooling rate of the molten alloy may increase with increasing frequency of vibration. For example, the vibration may lead to increased thermal contact between the molten alloy and the alumina mold. Fig. 10 showed, however, that the cooling condition was the same, irrespective of the vibration condition. Therefore, the decrease in secondary dendrites arm spacing resulted from an increase in the vibration-induced constitutional undercooling, rather than increased cooling rate in the mold.

Columnar dendrites incline toward the upstream direc-
tion when the liquid phase flows perpendicular to the growth direction of these dendrites.\textsuperscript{14–16} This results from the asymmetrical solutal field around the dendrite tip.\textsuperscript{17} The degree of constitutional undercooling on the upstream side is larger than that on the downstream side and hence, growth is enhanced in the upstream direction. Therefore, columnar dendrites grow more on the upstream side than on the downstream side, and become inclined during this enhanced growth. Furthermore, the secondary dendrite arm spacing near the tip decreases on the upstream side, owing to perturbation of the solid/liquid interface. This fluid-flow-induced change in morphology is the same as the aforementioned mechanism.

4.3. Complexity-determining Parameters

In this study, the fractal dimension was used to characterize the complexity of the equiaxed grains. The results revealed that the fractal dimension provides a suitable description of the morphology.

Figure 6 shows that, at a modifier content of 0.05 wt.%, the fractal dimension decreased initially and increased thereafter with increasing frequency of vibration. In contrast, the secondary dendrite arm spacing decreased monotonically with increasing frequency. These results indicate that the morphology becomes complex with increasing frequency of vibration. However, the fractal dimension provides only a partial description of the morphology of the solidified structure, and must therefore be used in conjunction with the secondary dendrite arm spacing \textit{etc.}, depending upon the situation.

The fractal dimension is the most appropriate parameter for describing the morphology of primary grains, when a solidified structure consists of globular equiaxed grains and secondary dendrite arms are difficult to define.

The shape of equiaxed grains may be estimated as a function of the solid fraction by using an etching technique or contours of the solute content. In this case, the morphology of the grains may be characterized in terms of the fractal dimension. The secondary dendrite arms are coarsened during solidification and therefore, estimating the secondary dendrite arm spacing as a function of the solid fraction is difficult.

5. Conclusions

The effect of mechanical vibration on the complexity of the solidified structure of an Al-10 wt.%Cu alloy was investigated. The conclusions of this study may be summarized as follows:

(1) Fluid flow at the solid/liquid interface, owing to mechanical vibration, may increase the complexity of the equiaxed grains.

(2) The size and complexity of the equiaxed grains can be independently controlled to some extent.

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REFERENCES