Effects of a high-gradient magnetic field on the migratory behavior of primary crystal silicon in hypereutectic Al–Si alloy

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Abstract
The migration of primary Si grains during the solidification of Al–18 wt%Si alloy under a high-gradient magnetic field has been investigated experimentally. It was found that under a gradient magnetic field, the primary Si grains migrated toward one end of the specimen, forming a Si-rich layer, and the thickness of the Si-rich layer increased with increasing magnetic flux density. No movement of Si grains was apparent under a magnetic field below 2.3 T. For magnetic fields above 6.6 T, however, the thickness of the Si-rich layer was almost constant. It was shown that the static field also played a role in impeding the movement of the grains. The primary Si grains were refined in the Si layer, even though the primary silicon grains were very dense. The effect of the magnetic flux density on the migratory behavior is discussed.

Keywords: high magnetic field, solidification, gradient magnetic field, Al-Si alloy, magnetization force, migration

(Some figures in this article are in colour only in the electronic version)

1. Introduction
It is well known that a magnetic field generates a magnetization force in materials, which may be used for magnetic separation [1, 2], magnetic levitation [3, 4], measuring the magnetic susceptibility of materials [5], and aligning the crystal orientation and texture of structures [6–8]. Because the magnetization force is proportional to the product of the field strength $B_z$ and its gradient $dB_z/dz$, for feeble magnetic materials, i.e. para- and diamagnetic substances, in a low magnetic field, the magnetization force is usually negligible because the values of magnetic susceptibility are only on the order of $10^{-4}$–$10^{-6}$. However, the utilization of a high magnetic field of several teslas makes it possible to visually observe the effect of magnetization forces on feeble magnetic materials. According to the difference in the susceptibilities of two substances, it is possible to separate them due to difference in the magnetization force. Sassa et al [9] reported that in the case of Al–Si alloy, the diamagnetic primary Si appeared to migrate in the paramagnetic liquid melt. In the case of no magnetic field, Si grains were mostly precipitated at the bottom of the melt, although some were precipitated on the side wall of the crucible. At the position of the positive magnetic field gradient, the Si grains were located at a lower region of the sample, which was similar to the case without a magnetic field, while the Si grains mostly segregated in the upper region of the sample at the position of the negative magnetic field gradient. We have previously reported a similar phenomenon of Si grain segregation in hypereutectic Al–Si alloy solidifying under a high-gradient magnetic field [10]. Wang et al found that the primary Si grain distribution was changed in a high magnetic field due to the action of
the Lorenz force and the magnetization force caused by the magnetic field on the migratory behavior of Si grains in molten aluminum [11]. Nevertheless, the product of the magnetic strength \( B_z \) and its gradient \( dB_z/dz \) has usually been regarded as a single factor when investigating the migration or segregation of primary silicon, and the individual effect of \( B_z \) and \( dB_z/dz \) has been ignored. It is interesting to determine the individual effect of \( B_z \) and \( dB_z/dz \), because the parameters may not only act as a driving force. Furthermore, the resistance force caused by the viscosity of the molten liquid has not been previously considered.

In this paper, the effects of magnetic flux density and its gradient on the migratory behavior of primary silicon in hypereutectic Al–Si alloy have been investigated separately, and a model was proposed to explain the mechanism of the observed behavior.

2. Experimental procedure

The experimental apparatus, as shown in figure 1, consists of a superconductor (SC) magnet, a furnace, a graphite crucible and a temperature monitor. A high static magnetic field can be produced with magnetic flux density up to 14 T in the center of the magnet with a gradient of the field away from the center. The distributions of the magnetic flux density and the parameter \( B_z dB_z/dz \) are shown in figure 2. The temperature inside the furnace could reach 900 °C, and was controlled by the temperature monitor with a precision of ±1 °C.

The Al–18 wt%Si alloy was prepared by melting pure Al and Si (with purities of 99.9 and 99.99%, respectively) in a resistance furnace. The melt was then poured into a graphite mold to cast specimens with a diameter of 9.5 mm and a length of 60 mm. The specimens were cut into 12-mm-long samples, and each sample was sealed in a graphite crucible during the solidification experiment, with the longitudinal axis in the vertical direction. The experimental procedure was as follows:

Experiment (1): the alloy sample was placed at positions in the field where the gradient of the field was maintained at about 25 T m\(^{-1}\) but with various magnetic flux densities, then was heated to 650 °C (in the mushy zone of the alloy) for 2.5 h, before being cooled to room temperature at a cooling speed of 15 °C min\(^{-1}\).

Experiment (2): the conditions were the same as those for experiment (1), except that the sample was placed at positions where the flux density was maintained at approximately 5 T with various magnetic field gradients.

Experiment (3): to examine the instantaneous solidification behavior of primary silicon, some samples were quenched from the mushy zone (at 650 °C).

The macro- and micro-solidification structures of longitudinal sections of the solidified samples were examined.

3. Experimental results

The results of experiment (1) are shown in figure 3. As can be seen from figures 3(a) and (b), which show similar macrostructures for the samples with 0 and 1.1 T magnetic flux density, the primary silicon grains mostly descended to the bottom of the samples, although some were precipitated
on the side walls. As the flux density increased to 2.3 T, the silicon grains appeared in the middle portion of the sample (see figure 3(c)) and the distribution of silicon grains became more uniform throughout the whole section. At a magnetic flux density of 6.6 T, the primary silicon grains migrated upward and formed a silicon-rich layer in the upper part of the specimen, as shown in figure 3(d). The layer maintained its thickness with further increase in the magnetic flux density to 7.7 and 9.9 T, as shown in figures 3(e) and (f), respectively. This phenomenon indicates that the effect of the magnetic flux density on the migration of primary silicon is to cause saturation when it reaches a critical value.

To determine the instantaneous solidifying behavior of the primary silicon, two samples were quenched from the mushy zone, and their macrostructures are shown in figure 4. As can be seen, the silicon-rich layer was formed before the solidification was completed in the sample solidified with magnetic field.

The macrostructures of samples with the same magnetic field strength and different gradients are shown in figure 5. One can see from this figure that the amount of separated primary silicon grains increased significantly with increasing magnetic field gradient. Note that the segregated primary silicon grains were refined gradually with increasing magnetic gradient.

To investigate the morphology of the silicon-rich layer, the microstructures of the sample with zero magnetic field (figure 3(a)), the segregated layers shown in figures 3(d)–(f) and the quenched samples (figures 4(a) and (b)) were examined. The results are shown in figure 6. Figures 6(a)–(f) correspond to the arrows labeled A–F in figures 3(a), (d), (e) and (f) and figures 4(a) and (b), respectively. From figure 6(a), it can be seen that the primary silicon grains became bulky and platelike at zero magnetic field, but the primary silicon grains in the segregated layers take on the shape of polygonal blocks, and the size of the grains is uniform under a gradient.
it can be deduced that the movement of the grain 

I

3

6

5

6

Cavity

B, C, D, E and F in figures

field, (a)–(f): microstructures corresponding to the arrows labeled A, mushy state, (f) quenching from the mushy state with zero magnetic (d) 9

(b) 5

Crystal silicon in hypereutectic Al–Si alloy under different magnetic (b), respectively.

Microstructures of the segregated layers of primary 

Figure 5. Macrostructures of Al–Si alloy solidified from 650 °C under magnetic field with various gradients. (a) 5.0 T, 12.4 T m⁻¹, (b) 5.1 T, 27.5 T m⁻¹, (c) 5.2 T, 38.0 T m⁻¹ and (d) 5.0 T, 44.7 T m⁻¹.

magnetic field, as shown in figures 6(b)–(d). The grains became smaller with increasing magnetic flux density. Upon comparing figures 6(d) and (e), it was found that the shape and size of the primary silicon grains were almost the same, even though the former solidified normally and the latter was quenched. However, the primary silicon grain size of the quenched sample with the magnetic field was smaller than that of the sample quenched without a magnetic field, as shown figures 6(e) and (f). In addition, upon comparing figure 3(e) (B₂ = 7.7 T, B₂ dB/ dz = 188 T² m⁻¹) with figure 5(c) (B₂ = 5.2 T, B₂ dB/ dz = 198 T² m⁻¹), and figure 3(f) (B₂ = 9.9 T, B₂ dB/ dz = 236 T² m⁻¹) with figure 5(d) (B₂ = 5.0 T, B₂ dB/ dz = 224 T² m⁻¹), it was found that for approximately the same magnitude of B₂ dB/ dz, i.e., the same magnetization force, the migration degree decreased with increasing B₂.

4. Discussion

In summary, the migratory characteristics of primary silicon under a gradient magnetic field can be described as follows:

Firstly, a silicon-rich layer was formed before the solidification was completed. Secondly, the effect of the magnetic flux density on the migration of primary silicon decreased once it reached a critical value. Thirdly, the amount of segregated primary silicon increased with increasing magnetic gradient. Fourthly, the grains in the silicon-rich layer were markedly refined with the formation of a segregated layer. Fifthly, the degree of refinement of the grains in the silicon-rich layer increased with increasing magnetic field gradient.

4.1. Effect of the magnetic flux density on the migration of primary silicon

The primary silicon accumulating in the upper portion of the samples was clearly due to the migration caused by the driving force of the magnetic field. To explain this thoroughly, the forces acting on a grain should be analyzed. As shown in figure 7, the forces acting on a grain include the effective gravitational force, the resistance force caused by the liquid viscosity, and the magnetic driving force proportional to B₂ dB/ dz. Also, a magnetic drag force is produced when the grain moves in liquid metal in a magnetic field. From figure 7 it can be deduced that the movement of the grain repels the melt with velocity Vₘ perpendicular to the direction of the magnetic field, inducing an induction current (I) in the melt. The current (namely, the conducting melt) interacts with the field to produce a Lorentz force f (defined as a unit area force) in the direction opposite the movement of the melt. In the upper portion, the motion of the grain behaves as if it is subjected to a resistance force Fᵣ, which we call a magnetic resistance force. In the lower part, the melt fills all
the available space due to its movement in front of the grain, but the Lorentz force tends to suppress this movement, and a drag force \( \mathbf{F}_{\text{D}} \), which acts on the grain, is generated, which we call the magnetic drag force. Thus, the net force on the grain is

\[
\mathbf{F} = \Delta \mathbf{F}_{\text{m}} + \Delta \mathbf{G} + \mathbf{F}_0 + \mathbf{F}_n + \mathbf{F}_{\text{D}} + \int \mathbf{f} \, ds,
\]

where \( \Delta \mathbf{F}_{\text{m}} \) and \( \Delta \mathbf{G} \) are the effective magnetization and gravitational forces, respectively.

On the basis of the principles of magnetization, \( \Delta \mathbf{F}_{\text{m}} \) can be written as

\[
\Delta \mathbf{F}_{\text{m}} = \nabla \times (\chi_1 \mathbf{H} + \chi_2 \mathbf{B}) \times \mathbf{B},
\]

where \( \mathbf{H} \) and \( \mathbf{B} \) are the magnetic field strength and the magnetic flux density, respectively, and \( \chi_1 \) and \( \chi_2 \) are the susceptibilities of the silicon and the surrounding liquid aluminum, respectively, and \( \mu_0 \) is the permeability of vacuum. \( \mathbf{B} \) is the magnetic field, \( \mathbf{G} \) is the gravitational field, and \( \mathbf{F}_{\text{D}} \) is the magnetic drag force, which is related to the shape and velocity of the primary silicon grain. Assuming the grain to be a sphere with radius \( r \), then, according to Stokes’ law, \( \mathbf{F}_n \) can be written as

\[
\mathbf{F}_n = 6 \pi r \eta \mathbf{v},
\]

where it is referred to as a characteristic length coefficient.

Generally, \( \mathbf{v} \) is proportional to \( \eta \); thus, equation (2) can be given as

\[
\int_S \mathbf{f} \cdot d\mathbf{s} = k' 6 \pi r V_d B_z^2,
\]

where \( k' \) is a coefficient related to the electrical conductivity of the melt and the radius of the grain.

Therefore

\[
F'_n = k' k'' 6 \pi r \mu_0 B^2 = k_0 6 \pi r \mu_0 B^2,
\]

where \( k'' \) is a ratio constant, and \( k_0 = k' k'' \), is a coefficient related to the electrical conductivity of the melt and the radius of the grain.

The magnetic drag force \( \mathbf{F}_{\text{D}} \) can be dealt with analogously. One can define \( \mathbf{F}_0 = \mathbf{F}_n + \mathbf{F}_{\text{D}} \), which we call the magnetic viscosity resistance force and can be written as

\[
F'_0 = k 6 \pi r \mu_0 B^2,
\]

where \( k \) is a ratio coefficient related to the electrical conductivity of the melt and the radius of the grain, which we also refer to as a characteristic length coefficient.

Substituting the above parameters and equation (5) into equation (1) results in

\[
F = \nabla \times (\chi_1 \mathbf{H} + \chi_2 \mathbf{B}) \times \mathbf{B} - V (\rho_1 - \rho_2) g - 6 \pi r \eta \left( \eta' + k B_z^2 \right).
\]

We define

\[
\eta' = \eta + k B_z^2,
\]

where \( \eta' \) is referred to as the effective dynamic viscosity of the melt. Using equation (6), the experimental results may be reasonably explained.

From equation (6), it is demonstrated that the grains do not move until the viscosity resistance force is overcome by the magnetization force (driving force); that is, the migration can take place only if the magnetization force exceeds a certain magnitude. Note that the fourth term in equation (6) is the resistance force caused by the Lorentz force, the magnetic viscosity force, which is, similarly to the magnetization force, proportional to the square of the magnetic flux density. Therefore, both the magnetic driving force and the magnetic viscosity force simultaneously increase with increasing magnetic flux density, and then counteract each other, resulting in a less rapid change in
the migratory behavior of the grains. Thus, the phenomenon shown in figures 3(d), (e) and (f) occurred, i.e. the thickness of the migratory layer was basically unchanged with increasing magnetic flux. On the other hand, for a constant magnetic flux, the driving force of the migration increases when the magnetic field gradient increases, thus resulting in increased segregation, as shown in figure 5. Further, from equation (6), for a constant magnetization force (i.e. the parameter \(B_z dB_z/dz\)), the resultant driving force decreases with increasing magnetic flux because the magnetic viscosity force is proportional to the square of the magnetic flux density, which reduces the migration of grains at higher densities. It is also predicted from equation (6) that the migration of the grains will be prevented if the magnetic flux density is sufficiently large.

4.2. Refinement of the primary silicon

In our previous paper, we reported the refinement of primary silicon under a high magnetic field [10]. We considered that one of the causes of refinement was related to the diamagnetism of the silicon. The probability of contact and collisions between the silicon grains decreased as a result of the diamagnetism because the grains repulsed each other, thus the distances among them were homogenous, resulting in the prevention of coarsening. However, in this paper it was found that for a constant magnetic flux, the primary silicon grains were refined when the magnetic gradient was increased. This indicates that the refinement of the grains is not only related to the repulsive force among them, but also to the magnetization force.

5. Conclusions

(1) A primary crystal silicon-rich layer was formed in hypereutectic Al–18 wt%Si alloy when it solidified from the mushy zone under a gradient magnetic field. The silicon-rich layer was formed before the solidification.

(2) At a constant magnetization force, the migratory degree of primary silicon grains decreased with increasing magnetic flux density.

(3) At a fixed magnetic flux density of 5 T, the amount of separated primary silicon grains increased with increasing magnetic field gradient, and the grain size simultaneously gradually decreased.

Acknowledgments

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