Electromagnetic Separation of Primary Crystallized $\text{Al}_{13}\text{Fe}_4$ Phase during Cooling Process of Molten Al–10%Fe Alloy*

Yosuke Tamura¹, Kentaro Mizuno² and Kenzo Takahashi²

¹Department of Advanced Materials Science and Engineering, Chiba Institute of Technology, Narashino 275-0016, Japan

²Zmag Ltd., Shiroy 270-1431, Japan

In this study, the electromagnetic separation behavior of primary $\text{Al}_{13}\text{Fe}_4$ crystallized in the cooling process of molten Al–10 mass%Fe alloy was clarified by observation of the solidification structure. The electromagnetic force was controlled by keeping the magnetic flux density constant and changing the applied direct current. The applied current was set in the range of 0 A to 100 A, and the experiment was conducted at intervals of 20 A to investigate the electromagnetic separation behavior of primary $\text{Al}_{13}\text{Fe}_4$.

During the cooling process, no separation effects of primary $\text{Al}_{13}\text{Fe}_4$ were observed, even when a current or magnetic field was independently applied. When both current and a magnetic field were applied, primary $\text{Al}_{13}\text{Fe}_4$ was miniaturized by the electromagnetic force and distributed in the surface layer of the ingot. However, no eutectic $\text{Al}_{13}\text{Fe}_4$ separation effect was observed. Even when the direction of the magnetic field was changed, primary $\text{Al}_{13}\text{Fe}_4$ was similarly distributed in the surface layer of the ingot. Electromagnetic separation effects were observed even in the range beyond the magnetic poles. When the length of the specimen was shortened to be within the range of the permanent magnet, no change was observed in the distribution behavior of primary $\text{Al}_{13}\text{Fe}_4$ in the surface layer of the ingot. During the cooling process, a negative pressure gradient was assumed to be formed from the specimen axis toward the surface, regardless of the direction of the electromagnetic force. [doi:10.2320/matertrans.F-M2019824]

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1. Introduction

Aluminum alloys are lightweight and increasingly being used for transportation equipment in order to improve fuel economy and, at the same time, more attention is being paid to the recycling of the used alloys.¹⁾ Aluminum smelting requires a considerable amount of electricity, while aluminum scraps, which have a high recycling rate, can be regenerated with only 3% of the energy required to manufacture a virgin ingot. However, the regenerated ingot must be diluted with a large amount of virgin ingot in order for it to be used as wrought materials.²⁾ Therefore, improving the quality of the regenerated ingot is essential in order to optimize energy savings in the life cycle of aluminum alloys.

Contamination is inevitable when remelting aluminum alloy scraps. When paints and foreign substances adhere to scraps they, along with alloy impurities, are mixed into the dissolution process, high melting point intermetallic compounds and insoluble nonmetallic inclusions are formed in the melt. These solid particles that intrude into the ingot impair the mechanical properties, workability, and corrosion resistance of the material, as well as causing unevenness during surface processing.³⁾ Therefore, the separation and removal of solid particles are very important, which has led to the studies of physical separation methods^{4–7)} such as gravity, centrifugal force and filtration of molten metal.

When the electrical conductivity between the molten metal and solid particles are different, it is possible to selectively separate solid particles using electromagnetic forces.^{8–11)} This method can separate fine particles over a relatively short time and the basic design of the equipment is simple, which has an advantage over the above mentioned methods. The theory of electromagnetic separation⁸⁾ has been known for many years

and in recent years, numerical analysis research has been conducted.^{12,13)} However, there are a limited number of cases that demonstrate its application to industrial materials and thus there is a lack of data to facilitate the implementation of this technology.

Contamination by iron impurities is unavoidable during melting of scraps. Iron has low solubility in molten aluminum and reacts with various elements to form high melting point Al–Fe intermetallic compounds, degrading the quality of regenerated ingots. Therefore, the separation and removal of impurities like iron is a very important issue.

In this study, the electromagnetic separation of primary $\text{Al}_{13}\text{Fe}_4$ crystallized during the cooling process of molten Al–10 mass%Fe alloy (hereinafter referred to as Al–10Fe) will be investigated by microstructural observation, and the study aims to obtain a fundamental understanding of the electromagnetic separation of iron impurities in the form of Al–Fe intermetallic compounds in aluminum alloys.

2. Experimental Aspects

2.1 Specimen and experimental procedure

Each specimen was an Al–10Fe alloy ingot machined into a round bar with a diameter of 18 mm and a length of 90 mm, which weighed approximately 66 g. Each specimen was inserted into a 120 mm long mullite tube with an inner diameter of 20 mm, as shown in Fig. 1, and each end was plugged with a graphite electrode. The side of each electrode facing the specimen was made into a pointed profile; as a result, the oxide film covering the specimen surface would most likely be broken when the specimen melted and the current supply to the molten metal would be stabilized. A copper plate with a thickness of 5 mm and a width of 50 mm was fastened to each graphite electrode with stainless steel bolts and nuts. This assembly was placed in an electric furnace maintained at 1000°C and the specimen was melted.

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Fig. 1 Schematic diagram of specimen assembly, showing Al-10Fe alloy inserted into a mullite tube and closed at each end with a graphite electrode.

Fig. 2 Direction of magnetic flux density B , current density J , and generated electromagnetic force density F (downward) in relation with specimen.

It was then removed from the furnace and the molten metal therein was cooled at room temperature under one of the following conditions:

- (i) Neither current nor magnetic field applied
- (ii) Current only applied
- (iii) Magnetic field only applied
- (iv) Both current and magnetic field applied

For conditions (iii) and (iv), the specimens were immediately set between the magnetic poles of neodymium magnets after they were taken from the furnace. The neodymium magnets were 50 mm long, 50 mm wide, and 14 mm thick. An insulator sheet was placed between the magnet and specimen to protect the magnet surface from heat. The magnetic pole interval was 30 mm and the magnetic flux density between the magnetic poles was confirmed to be 0.54 T.

In condition (iv), the current density J , the magnetic flux density B , and the electromagnetic force density $F (= J \times B)$ were arranged as shown in Fig. 2; these are all vector quantities. Figure 3 is a schematic diagram of the experimental set-up. In order to measure the temperature, a sheathed thermocouple was inserted through a hole drilled in the center of the mullite tube.

The Al-Fe phase diagram¹⁴⁾ is shown in Fig. 4. The current and/or magnetic field for conditions (ii)–(iv) were applied while the specimens were still approximately 1000°C. The solidification of Al-10Fe starts when the temperature drops to approximately 880°C (Fig. 4), which means that the experimental conditions were applied before the primary crystal, $\text{Al}_{13}\text{Fe}_4$, began to nucleate.

Optical microscope, scanning electron microscope (SEM), and an electron probe microanalyzer (EPMA) were used for microstructural observation and analysis. The acceleration voltages used for SEM and EPMA were both 15 kV.

2.2 Theoretical considerations for determination of applied current

When a current and magnetic field are applied to molten

Fig. 3 Schematic illustration of experimental set-up for applying magnetic field and current as shown in Fig. 2.

Fig. 4 Phase diagram of the Al-Fe system.¹⁴⁾

metal to satisfy the relationship in Fig. 2, the electromagnetic force density $F (= J \times B)$ acts on the molten metal as a vector product of the current density J and the magnetic flux density B . Furthermore, when coordinates with vertical depth direction z relative to the surface of the molten metal are adopted, the following relationship is obtained:

$$\frac{dp}{dz} = \rho_L g + |J \times B| \quad (1)$$

Where dp/dz is the pressure gradient, ρ_L is the density of the molten metal, and g is the gravitational acceleration. Although $\rho_L g$ is constant in eq. (1), the magnitude of electromagnetic force density $|J \times B|$ can be controlled by changing the applied current.

When a pressure gradient is formed in molten metal, the force $\rho_L g V + G_c V |J \times B|$ acts upward on the non-conductive particles having volume V . In this equation, $\rho_L g V$ is buoyancy, $G_c V |J \times B|$ corresponds to the surface force exerted to the particle surface by molten metal, and G_c is a constant related to the distribution disturbance of the current density in the vicinity of particles.¹²⁾ As an example of G_c , if the particles are spheres, G_c is 3/4.

When particles of density ρ_p are moving downward relative to the molten surface, drag D acts on the particles in the opposite, upward direction. If the particles are

considered to be spheres, the magnitude of gravity acting on the particles is $\rho_L g V$, and the force acting upward is positive, the following equation of motion can be established:¹³⁾

$$\rho_p V \left(\frac{du}{dt} \right) = \rho_L g V + \frac{3}{4} V |\mathbf{J} \times \mathbf{B}| + D - \rho_p g V \quad (2)$$

The du/dt term on the left side represents the acceleration of the particles and $D = 0$ if the particles that crystallize from the molten metal are regarded as a stationary state. Therefore, if the following equation (3) is satisfied, the non-conductive particles accelerate upwards along the vertical direction.

$$\begin{aligned} \rho_L g V + \frac{3}{4} V |\mathbf{J} \times \mathbf{B}| - \rho_p g V &> 0 \\ \Leftrightarrow |\mathbf{J} \times \mathbf{B}| &> \frac{4}{3} (\rho_p - \rho_L) g \end{aligned} \quad (3)$$

In the case of conductive particles, the relationship between the electric conductivity σ_L of molten metal and the electric conductivity σ_p of solid particles must be considered. Generally, the impurity particles are nonmetallic inclusions and intermetallic compounds; therefore, we can assume that $\sigma_L > \sigma_p$. In the case of conductive particles, a body force develops within particles; hence, based on the theory of Leenov *et al.*,⁸⁾ eq. (2) is rewritten as follows:

$$\begin{aligned} \rho_p V \left(\frac{du}{dt} \right) &= \rho_L g V + \left(\frac{-3\sigma_p}{2\sigma_L + \sigma_p} \right) V |\mathbf{J} \times \mathbf{B}| \\ &+ \left\{ 1 - \frac{1}{2} \left(\frac{\sigma_L - \sigma_p}{2\sigma_L + \sigma_p} \right) \right\} V |\mathbf{J} \times \mathbf{B}| + D - \rho_p g V \end{aligned} \quad (4)$$

On the right side of eq. (4), the second term corresponds to the body force and the third term corresponds to the magnitude of the surface force. Here, the body force and the surface force always act downward and upward, respectively. For conductive particles, eq. (3) can be rewritten as eq. (5):

$$|\mathbf{J} \times \mathbf{B}| > \frac{2}{3} f (\rho_p - \rho_L) g \quad (5)$$

Furthermore,

$$f = \frac{2 + \left(\frac{\sigma_p}{\sigma_L} \right)}{1 - \left(\frac{\sigma_p}{\sigma_L} \right)} \quad (6)$$

Equation (6) is a fractional function and as shown in Fig. 5, the curve has an asymptote at $x = 1$ in the range of $\sigma_L > \sigma_p$. Therefore, f rapidly increases beyond $\sigma_p/\sigma_L \sim 0.8$, which implies that it becomes difficult to separate particles. However, since the rate of increase of f is relatively small for $\sigma_p/\sigma_L < 0.8$, the force required to separate particles can be estimated if we assume that the particles are nonconductive.

The intermetallic compound of interest in this study is Al₁₃Fe₄. Equation (7) is obtained by substituting the densities of Al₁₃Fe₄ ($\rho_p = 3.84 \text{ Mg/m}^3$)¹⁵⁾ and molten aluminum ($\rho_L = 2.39 \text{ Mg/m}^3$) into eq. (5):

$$|\mathbf{J} \times \mathbf{B}| > 9.5f \quad (7)$$

The intermetallic compound Al₁₃Fe₄ is monoclinic and its electrical resistivity varies with axial direction;¹⁶⁾ existing data include electrical resistivity values up to room temper-

Fig. 5 Graphical representation of eq. (6); σ_L and σ_p are the electrical conductivities of molten metal and solid particles, respectively.

ature, but the temperature coefficient is positive. The value of electrical conductivity of Al₁₃Fe₄, when converted from the values of electrical resistivity of Al₁₃Fe₄ at room temperature, ranges from 0.17×10^6 to $1.0 \times 10^6 \Omega^{-1} \text{ m}^{-1}$, whereas the electrical conductivity of aluminum is $38 \times 10^6 \Omega^{-1} \text{ m}^{-1}$ at room temperature and $4.0 \times 10^6 \Omega^{-1} \text{ m}^{-1}$ just above its melting point.¹⁷⁾ Therefore, assuming $\sigma_p/\sigma_L \approx 0$, the initial condition of eq. (7) was $|\mathbf{J} \times \mathbf{B}| > 19 \text{ kN/m}^3$, which can be transformed to $|\mathbf{J} \times \mathbf{B}| > 2.1 \times 10^3 I$ where I is the current by using the initial specimen cross-sectional area. According to this relationship, an applied current greater than 9 A is required to separate primary Al₁₃Fe₄ (hereinafter referred to as the primary crystals) from molten aluminum. Based on this threshold, experiments were conducted with applied current set to 0, 20, 40, 60, 80, and 100 A.

3. Results

3.1 Effects of current and magnetic field

Figure 6 shows the macrostructure of the specimen (hereinafter referred to as the ingot) obtained under condition (i). Needle-like phases, indicated by an arrow in the figure, were clearly observed in the structure; based on the Al–Fe phase diagram (Fig. 4), this phase corresponds to the primary crystal.

Figure 7(a) displays the macrostructure of the ingot obtained under condition (ii) with applied current of 100 A, and Fig. 7(b) shows the macrostructure of the ingot obtained

Fig. 6 Macrostructure of transverse cross-section of ingot. Neither current nor magnetic field was applied. The needle-like phase indicated by an arrow is primary Al₁₃Fe₄.

Fig. 7 Macrostructure of transverse cross-section of ingots obtained with application of either (a) current of 100 A DC or (b) magnetic field alone during cooling.

Fig. 8 Images of ingots obtained under: (a) no magnetic field and no current and (b) both magnetic field and current (100 A DC).

under condition (iii). When either current (ii) or magnetic field (iii) was applied alone, the size and shape of the primary crystals were not markedly different from those in Fig. 6 and no changes were observed in the distribution.

3.2 Effect of electromagnetic force

Figure 8 shows photographs of the ingots (a) and (b) obtained under conditions (i) and (iv), which correspond to applied currents equal to 0 and 100 A, respectively. Under condition (iv), a part of the ingot (b), that was in-between the magnetic poles, is heavily recessed and the sides appear to have bulged.

Figure 9 shows the cross-sectional macrostructures that were taken at the location 10 mm from the electrode tip (see Fig. 1). These images illustrate the change in solidification structure with increasing current. The location was chosen because each ingot has the same circular cross-sectional shape and no defects, such as cavities, were observed at this position. Moreover, based on observations from the entire length of the ingot, it was determined that 10 mm from the electrode tip was a suitable location for observing the distribution of $\text{Al}_{13}\text{Fe}_4$. Comparing each structure, the primary crystals are clearly refined and distributed toward the outer periphery of the ingot above 40 A, and above 60 A the tendency is much clearer for the ingots to exhibit a distinctive structure, in that the primary crystals are distributed along the concentric circle near the surface of the ingot. Such structures are convenient for removing primary crystals from the ingot and are intriguing from the point of view of surface modifications, development of composite materials, etc.^{10,18,19)}

Fig. 9 Transverse cross-section of ingots, showing the change in solidification structure with applied current.

Fig. 10 EPMA line analysis for iron across the diameter of cross-sections in Fig. 9.

The results of EPMA line analyses for iron are shown in Fig. 10. The analyzed diameter line was chosen randomly. When the applied current reached or exceeded 40 A, there were sections of the ingot cross-section where the across the diameter of the cross-sections in Fig. 9 iron content reached a minimum level shown by an arrow in Fig. 10. These regions of constant low iron content corresponded to the areas of the eutectic region without any primary crystals, shown in Fig. 11, where the white phase is eutectic $\text{Al}_{13}\text{Fe}_4$ and the dark phase is eutectic Al. Comparing the level of low iron regions in Fig. 10 shows that it is unaffected by the magnitude of the applied current, and thus it would be difficult to separate the eutectic $\text{Al}_{13}\text{Fe}_4$ from the aluminum even with an applied electromagnetic force.

Figure 12 shows an example of a cooling curve when both current (100 A) and magnetic field were applied. It takes approximately 3 minutes from the time ingot was set between

Fig. 11 Back-scattered SEM image of eutectic structure, observed in an area where the iron content was constant.

Fig. 12 Cooling curve when both magnetic field and current (100 A DC) were applied.

Fig. 13 Direction of magnetic flux density B , current density J , and generated electromagnetic force density F (horizontal direction) in relation with specimen.

Fig. 14 Image of an ingot after both a magnetic field and current (100 A DC) were applied to meet the relationship shown in Fig. 13. Lines ① to ⑨ indicate the locations for observing the transverse cross-section of solidification structures.

the magnetic poles until the temperature cooled to the eutectic temperature during which separation of the primary crystals would take place.

3.3 Effect of direction and range of electromagnetic force

The specimen was placed between the magnetic poles to satisfy the conditions shown in Fig. 13 with an applied current of 100 A. Figure 14 is an image of the ingot obtained; the part of the ingot located between the magnetic poles was heavily depressed. Figure 15 shows the macrostructure of cross-sections cut from the ingot at 10 mm intervals (①–⑨ in the figure). In all cross-sections, except for cross-section ⑨ which was near the graphite electrode, the primary crystals, as observed previously (Fig. 9), were distributed near the surface of the ingot and the distribution of the primary crystals was consistent even in positions ①, ②, and ⑧ that were located outside the magnet.

The experiment was also conducted with an ingot that was shortened from 90 mm to 50 mm in order for the entire ingot to fit within the magnetic poles. The applied current was 100 A. Since the more uniform magnetic field was applied to the entire ingot, the flow of the molten metal, arising from non-uniformity in the electromagnetic force as in the 90 mm ingot, would be suppressed. Figure 16 shows the longitudinal

Fig. 15 Macrostructure of transverse cross-sections of ingot at locations ①–⑨ in Fig. 14, showing the distribution of the primary $\text{Al}_{13}\text{Fe}_4$ crystals.

Fig. 16 Longitudinal sections of the ingots obtained with (bottom image) and without the electromagnetic force (top image), showing the distribution of primary crystals. Note that the left end is the surface that was in contact with the graphite electrode and the right end of the image is the center of the ingot. The black area observed in ingot (bottom image) is a cavity.

sections of the resultant ingots with and without the electromagnetic force. As in Fig. 9 and 15, with the electromagnetic force, the primary crystals were distributed near the concentric surface of the ingot (Fig. 16 bottom image), and by observing the end section of the ingot, the surface of which was in contact with the graphite electrode was also found to be covered with primary crystals. This is a potential explanation of why primary crystals were observed over the entire cross-section of ⑨ in Fig. 15.

4. Discussion

4.1 Electromagnetic separation behavior

The results of this study clearly demonstrated the effectiveness of electromagnetic separation of primary $\text{Al}_{13}\text{Fe}_4$ crystals. However, the observed distribution of primary crystals differs from the distribution expected from theory. The results show that on the contrary to the expected idea that the primary crystals would be segregated at the upper part of the ingot, they were uniformly distributed around the ingot surface. For electromagnetic separation using DC current and a static magnetic field, such a structure is difficult to explain. Park *et al.*¹⁸⁾ reported that primary silicon crystallized on the ingot surface when a hyper-eutectic Al–Si alloy was melted in a quartz tube and solidified while applying electromagnetic force. They concluded that silicon crystals were formed by the reduction of the quartz tube (SiO_2) with molten aluminum. However in the present study, there is almost no chance for iron to be introduced from the mullite tube.

The ingots shown in Figs. 8(b) and 14 suggest that the electromagnetic force acted during the cooling process, but the directions of deformation (depression) and the electromagnetic force did not always coincide, and it is not clear during which stage of the cooling process depression occurred. Therefore, it is difficult to determine the direction

of the electromagnetic force from the appearance of the ingot. However, primary crystals are expected to gather at places where the molten metal pressure is small; therefore, the solidification structures shown in Figs. 9, 15, and 16 suggest that a negative pressure gradient formed from the central axis of the molten metal toward the surface layer.

According to the Biot-Savart Law, when an electric current is applied, a magnetic field is formed in the clockwise direction as the current travels away down the axis. Therefore, if the magnetic field induced by the current exerts a considerable influence, the electromagnetic force is expected to act from the radial direction toward the center in the presence of a current (Pinch effect) and reach a state close to the pressure gradient as described above. However, as shown in Fig. 7(a), the magnetic field generated solely by the applied current is not large enough to affect the solidified structure.

In order to consider these results in terms of electromagnetism, the distribution of current density and magnetic flux density in the molten metal, and the interaction between the external magnetic field and magnetic field induced by current must be investigated further.

4.2 Formation of solidified structure

Regardless of experimental conditions (i) to (iv), nucleation of primary crystals occurs on the cooling surface of molten metal. Assuming the above-mentioned pressure gradient exists under condition (iv), it is possible that the primary crystals, nucleated on the molten ingot surface, were prevented from moving into the higher pressure regions of the ingot. In subsequent solidification processes, a eutectic grew from the surface and also from the primary crystals if $\text{Al}_{13}\text{Fe}_4$ is the leading phase of the eutectic. Hence, a solidified shell in which primary crystals were captured was first formed in the beginning of solidification, followed by the eutectic growth to the interior of ingot, resulting in the solidified structure as shown previously (Figs. 9 and 14). Based on this concept, an electromagnetic field and current should be applied before nucleation begins in order to obtain effective separation by restricting the dispersion of the primary crystals.

When setting the experimental conditions, the shape of the primary crystals was assumed to be spherical; however, the primary crystals were determined to be needle-like, as shown in Fig. 6. Therefore, primary crystals were exposed to lateral forces during the growth process. Since these forces were distributed across the length of the primary crystals, the primary crystals were likely crushed by forces like rotation or collision. As a result of these forces, the primary crystals appear to have been refined, as shown in Fig. 9.

5. Conclusions

Primary $\text{Al}_{13}\text{Fe}_4$ in molten Al–10Fe alloy was electromagnetically separated under predetermined conditions based on existing theory. However, the distribution of primary crystals after electromagnetic separation was not biased to one side of the ingot as expected by the theory. Instead the primary crystals were distributed uniformly near the surface of the specimens; thus, ingots covered with primary crystals

were obtained. Although the present study does not investigate these phenomena in terms of electromagnetism, the results offer new information about the electromagnetic separation of Al–Fe intermetallic compounds from molten aluminum.

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