Investigations on the Solidification Behavior of Al-Fe-Si Alloy in an Alternating Magnetic Field

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Effects of a low frequency alternating magnetic field on the solidification behavior of the Al-Fe-Si alloy were investigated. Solidification characteristics of the alloy were predicted by software Thermo-Calc and compared to the experimental observations. The solidification sequences of the alloy solidified with and without the application of the AC magnetic field were confirmed by DSC and structural measurements. Al₃Fe was the dominant phase in the alloy. A small amount of α-AlFeSi phase with its characteristic dendritic or Chinese script-like morphology was observed to grow in close association with Al₃Fe. Distribution of Al₃Fe phase was almost homogeneous in the volume of the sample when the alloy was solidified in the conventional condition. When the AC magnetic field was imposed, the Al₃Fe phase was accumulated towards the center of the sample. Macrohardness profiles were in good agreement with the structural observations. [doi:10.2320/matertrans.47.2092]

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1. Introduction

Iron and silicon are the most common impurities in aluminium base alloys. Because the solid solubility of iron in aluminium is less than 0.05 mass% at equilibrium, almost all the iron in aluminium alloys forms intermetallic secondary phases.¹² A large number of iron-containing intermetallic phases have been identified in the microstructures, depending on the solidification conditions and alloy composition. Binary Al-Fe phases predominate in low Si alloys or high Fe alloys, including equilibrium Al₃Fe and metastable Al₀.₇Fe, Al₀.₉Fe₃, Al₁.₃Fe and Al₁.₂Fe, respectively. The formation of ternary Al-Fe-Si phases is available when Si content is relatively similar to the Fe content of the alloy, such as α (Al₀.₃Fe₀.₇Si or Al₀.₇Fe₂Si₀.₃), β (Al₁.₃FeSi or Al₁.₂Fe₂Si₀.₃) and δ (Al₁.₂FeSi₀.₃).¹⁻⁷ These particles influence the material properties during subsequent fabrication steps and play a crucial role for the material quality. Among them, the needlelike Al₃Fe and β phases were reported to be deleterious to the mechanical properties, especially, particles of primary crystals, which represent strong stress concentrators and promote the initiation of sharp microcracks. The growth of microcracks may cause brittle fracture. On the other hand, the sharp edges of these phases are responsible for the reduction of ductility and toughness.³⁻⁵³ Thus, it is of considerable technological interest to control the morphology and distribution of these phases in order to eliminate the negative effects mentioned above.

So far, a lot of studies have been devoted to the neutralization of the harmful influence of iron-containing phases. Addition of such elements as Mn, Be, Cr, Co or Ni can favor an increase inductility of aluminium alloys by modifying the needlelike morphology of iron-containing phases to a less harmful, more compact form.³¹₀⁻¹₂ Other methods including the melt superheating,¹³ heat treatment,¹⁰ increasing cooling rate,¹⁵ magnetic field significantly modified the flow pattern and temperature field in the sump, and these effects led to a substantial amelioration of the surface quality and a marked reduction of the grain size. Furthermore, the solubility of alloying elements was effectively promoted and the macrosegregation was reduced, especially the possibility of hot cracking in the production of large size ingot was markedly reduced.¹²³ However, the effects of the low frequency magnetic field on the iron-containing intermetallic phases formed during solidification have not been specially explored.

The present investigation was hence aimed at studying the morphology and distribution of the iron-containing intermetallics in the Al-Fe-Si alloy under an applied low frequency alternating (AC) magnetic field. Attempts are also made to predict thermodynamically the solidification path of the alloy. Such prediction is then compared to the experimental results.

2. Experimental

2.1 Sample preparation

The chemical compositions of the Al-Fe-Si alloy investigated in the present study are given in Table 1. The alloy was prepared from high-purity aluminium, iron, and silicon to avoid any contamination. The preparation was carried out in the laboratory medium-frequency induction furnace with a graphite crucible, details of which have been described elsewhere.¹⁵⁻²⁰

The equipment used in the present study is schematically...
shown in Fig. 1. It consists of an electrical resistance furnace and a 380 turns water-cooled copper coil. The specimens, 35 g, were cut from the alloy ingots, homogenized and then heated to 680°C in corundum crucibles in the furnace. In order to melt the material entirely and homogenize the composition, the melt was kept for 20 minutes at that temperature, and then cooled in the furnace at a cooling rate of 0.05°C/s with the simultaneous application of the AC magnetic field until the temperature decreased to room temperature. Throughout the experiment, the coil electrical current was maintained at 350 A with the exciting frequency of 20 Hz, while the magnetic flux density at the center of the current was maintained at 350 A with the exciting frequency of 20 Hz, while the magnetic flux density at the center of the coil; (5) coil; (6) corundum crucible.

### Table 1 Compositions of the Al-Fe-Si alloy used in the present work.

<table>
<thead>
<tr>
<th>Element (mass %)</th>
<th>Fe</th>
<th>Si</th>
<th>Ca</th>
<th>Ti</th>
<th>Cu</th>
<th>Pb</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1.185</td>
<td>0.495</td>
<td>&lt;0.00003</td>
<td>&lt;0.00005</td>
<td>&lt;0.0002</td>
<td>&lt;0.00005</td>
<td>Bal.</td>
</tr>
</tbody>
</table>

In the Thermo-Calc’s User Guide. In addition to the equilibrium solidification model, the Gulliver-Scheil model, which neglects any solid-state diffusion, was mostly used in calculation. The software was firstly applied to determine the phases crystallized and their amount as well as crystallization temperatures. Secondly, effects of the content of iron and silicon on the amount of major phases and their crystallization temperatures were investigated. Such thermodynamic prediction was compared to the experimental results.

### 2.3 DSC measurements

The solidification behavior of the Al-Fe-Si alloy was investigated by differential scanning calorimeter (DSC) analysis. The flat disc-shaped DSC specimens (0.5 mm thick and 2 mm in diameter) were removed from the alloy samples. Very small specimens (4–8 mg) were used to get a fast response, minimum thermal lag, high peak resolution and temperature accuracy. The specimens were cleaned in an ultrasonic cleaner and the measurements were carried out using a TA DSC-Q100 system operating under dynamic nitrogen atmosphere. Placing a specimen in the sample cell and an empty alumina crucible in the reference cell at 50°C, the calorimeter was quickly heated up (at a heating rate of 3.5°C/s) to ~690°C, held isothermally at this temperature for 20 minutes, cooled at 0.05°C/s over the temperature range 690–500°C, and finally cooled to room temperature.

### 2.4 Sample characterization

A comparative metallographic study was made on samples solidified with and without the application of the AC magnetic field. Each sample was sectioned vertically and prepared using a standard procedure for the metallographic examination. The microstructures measurements were made by the optical microscopy Leica DMR. The chemical compositions of iron-containing intermetallics were determined by using a SHIMADZU SSX-550 electron probe microanalysis (EPMA) operated at an accelerating potential of 15 kV. To determine the difference in hardness between edge and center of the sample, Vickers hardness was measured with 3-Kg load and 10-s dwell time in a digital macrohardness tester (450SVD). The testing was carried out on polished samples at an increment of 2 mm across the diameter.

### 3. Results

#### 3.1 Solidification characteristics

The variation of calculated fraction solid with crystallization temperature in the Al-Fe-Si alloy is shown in Fig. 2. The solidification paths calculated from the equilibrium and Gulliver-Scheil solidification models are almost identical except the temperatures at which solidification completes. In the case of the equilibrium solidification model, the aluminum FCC starts to crystallize at 653.58°C. As the temperature falls, the liquid becomes more and more enriched in Fe and Si, resulting finally in the interdendritic liquid regions from which Al<sub>3</sub>Fe and α-AlFeSi precipitate at 649.71°C and 630.56°C, respectively. However, from the results calculated from the Gulliver-Scheil model, in addition to Al<sub>3</sub>Fe and α-AlFeSi, β-AlFeSi and Si appear near the end of solidification.
Figure 3 shows the typical DSC traces of samples solidified with and without the magnetic field. In the DSC curves, the reaction peaks reflect the specific phase changes and the peak area is proportional to the heat of reaction associated with the phase transformation. As shown in Fig. 3, two reaction peaks were identified from the DSC curves of this Al-Fe-Si alloy solidified in two conditions. Peak 1 corresponds to the development of aluminum dendrites; peak 2 represents the main binary eutectic reaction, forming the \( \text{Al}_3\text{Fe} \) phase.

A comparative evaluation of crystallization temperatures obtained via the phase diagram calculations by the Gulliver-Scheil model and DSC measurements is given in Table 2. Table 2 also lists the weight percentage of each phase at complete solidification temperature. A good agreement is observed between the calculated and measured values. Unlike the \( \text{Al}_3\text{Fe} \) phase, the heats of formation of the \( \beta\text{-AlFeSi} \) phase observed in the sample were too weak to be detected due to their small amount.

### 3.2 Microstructure

Figure 4 shows the as-cast microstructures of the Al-Fe-Si alloy solidified in two solidification conditions. It can be seen that needle-like \( \text{Al}_3\text{Fe} \) phase dominated the microstructure at the grain boundaries, forming an almost continuous network. In Figs. 4(a) and (b), the distribution of \( \text{Al}_3\text{Fe} \) phase was almost homogeneous in the volume of the sample, and there were no remarkable differences in the average grain size between the edge and the center of the sample when the alloy was solidified in the conventional condition. However, when the AC magnetic field was imposed, the amount of \( \text{Al}_3\text{Fe} \) phase decreased obviously at the edge of the sample, while the center exhibited much higher volume fraction of \( \text{Al}_3\text{Fe} \) phase than the edge did, as shown in Figs. 4(c) and (d). It suggests that \( \text{Al}_3\text{Fe} \) phase tends to accumulate towards the center by imposition of the AC magnetic field.

The optical micrographs in Fig. 5 provide further information about the microstructures, which consisted of two intermetallic compounds: \( \text{Al}_3\text{Fe} \) and \( \beta\text{-AlFeSi} \). They were identified by a combination of morphology and EPMA analysis. The \( \text{Al}_3\text{Fe} \) phase was the dominant phase in two solidification conditions. A small amount of the \( \alpha\text{-AlFeSi} \) phase with its characteristic dendritic or Chinese script-like morphology was observed to grow in close association with the \( \text{Al}_3\text{Fe} \) phase. The average composition of the \( \alpha\text{-AlFeSi} \) phase was 59.02 mass% Al, 32.69 mass% Fe, and 8.29 mass% Si, corresponding to a formula of \( \text{Al}_{1.4}\text{Fe}_2\text{Si} \).

### 3.3 Macrohardness

The distribution of hardness across the transverse section of the sample is an important factor determining the effects of the AC magnetic field on the as-cast structure. Figure 6 shows the Vickers macrohardness profiles measured at an increment of 2 mm along the diameter. For the sample solidified in the AC magnetic field, the flow curve shows a sharp hardness peak at the sample center and a significant
decrease at the regions close to the sample edge. However, the hardness shows no distinctive difference between the edge and the center of the sample when the alloy was solidified in the conventional condition.

4. Discussions

4.1 Solidification sequence of the Al-Fe-Si alloy

The formation of the intermetallic phases during solidification sequence of the Al-Fe-Si alloy is illustrated in Fig. 4 and Fig. 5. Fig. 4 shows optical micrographs of the Al-Fe-Si alloy: (a) edge and (b) center of the sample solidified without the AC magnetic field; (c) edge and (d) center of the sample solidified with the AC magnetic field. Fig. 5 presents optical micrographs of the samples solidified (a) without and (b) with the AC magnetic field, illustrating the close association observed between $\text{Al}_3\text{Fe}$ and $\alpha$-$\text{AlFeSi}$; (c) and (d) higher magnification micrographs of areas outlined by frames in (a) and (b), respectively.
The solidification of dilute Al-Fe-Si alloys has been reported to take place through the following reactions:\(^{6,27–29}\)

\[
\begin{align*}
L & \rightarrow \alpha-\text{Al} \\
L & \rightarrow \alpha-\text{Al} + \text{Al}_3\text{Fe} \\
L + \text{Al}_3\text{Fe} & \rightarrow \alpha-\text{Al} + \alpha-\text{AlFeSi} \\
L & \rightarrow \alpha-\text{Al} + \alpha-\text{AlFeSi} \\
L + \alpha-\text{AlFeSi} & \rightarrow \alpha-\text{Al} + \beta-\text{AlFeSi} \\
L & \rightarrow \alpha-\text{Al} + \beta-\text{AlFeSi} \\
L & \rightarrow \alpha-\text{Al} + \beta-\text{AlFeSi} + \text{Si}
\end{align*}
\]

In the initial stage, the primary aluminum dendritic phase nucleates and grows. As the temperature decreased, the \(\text{Al}_3\text{Fe}\) binary eutectic reaction (2) takes place. Subsequently, \(\text{Al}_3\text{Fe}\) is transformed into \(\alpha-\text{AlFeSi}\) by peritectic reaction (3).

On further cooling, \(\alpha-\text{AlFeSi}\) forms through a eutectic reaction (4) simultaneously with aluminum and then transforms into \(\beta-\text{AlFeSi}\) by peritectic reaction (5). The last stages are the binary eutectic reaction (6) producing Al and \(\beta-\text{AlFeSi}\), and finally a ternary eutectic reaction (7) giving Si in addition to the previous two phases.

The type of the intermetallic phases present in the solidification microstructure and the temperatures at which they crystallize primarily depend on alloy composition. In the present study, effects of the content of iron and silicon on the solidification characteristics of the Al-Fe-Si alloy were also calculated by the Gulliver-Scheil model. The relationships between the calculated crystallization temperatures and predicted amount of intermetallics with iron are shown in Figs. 7(a) and (b). With increasing iron content, the crystallization temperatures of aluminum FCC decreases slightly while those of Si remain almost constant. The \(\text{Al}_3\text{Fe}\) temperature increases moderately at 0.22–1.96 mass% and then rises up substantially and linearly with iron content. The crystallization temperatures of other two phases, \(\alpha-\text{AlFeSi}\) and \(\beta-\text{AlFeSi}\), increase at first and then stay at 630.56 and 611.78°C, respectively. It is clear that the amount of \(\text{Al}_3\text{Fe}\) is very sensitive to iron content. It does not crystallize until the iron content is added up to 0.22 mass% and increases significantly with iron. \(\alpha-\text{AlFeSi}\) starts to crystallize at about 0.085 mass% Fe. The quantity of \(\alpha-\text{AlFeSi}\) increases greatly in the range of 0.085 to 2.2 mass% of iron and stays at about 0.38 mass%. Similar to the case of \(\alpha-\text{AlFeSi}\), \(\beta-\text{AlFeSi}\) increases linearly from 0.005 to 0.085 mass% Fe and remains almost constant after that. The amount of Si decreases moderately at first and then has the same tendency as that for \(\beta-\text{AlFeSi}\).

Figure 8(a) shows the relationship between the crystallization temperatures of intermetallic phases and silicon content. The crystallization temperatures of aluminum FCC and \(\text{Al}_3\text{Fe}\) go lower slightly with increasing silicon content while those of \(\beta-\text{AlFeSi}\) and Si remain almost constant. The \(\alpha-\text{AlFeSi}\) temperature negligibly changes at first but gradually decreases from 2.4 mass% Si. Beyond 2.4 mass% Si, \(\text{Al}_3\text{Fe}\) does not crystallize any more. The variation of the predicted amount of intermetallic phases with silicon is shown in Fig. 8(b). The quantity of \(\text{Al}_3\text{Fe}\) decreases greatly in the range of 0.0 to 2.4 mass% of silicon and decline moderately at 2.4–3.0 mass%. \(\alpha-\text{AlFeSi}\) starts to crystallize at about 0.1 mass% Si. With increasing silicon content, the amount of \(\alpha-\text{AlFeSi}\) rises up remarkably at silicon content 0.1–2.4 mass% and then goes down significantly at 2.4–5.4 mass%. It is clear that the amount of \(\text{Al}_3\text{Fe}\) and \(\alpha-\text{AlFeSi}\) goes to zero at 3.0 and 5.4 mass%, respectively. This is attributed to the completion of peritectic reactions (3) and (5). \(\beta-\text{AlFeSi}\) and Si start to crystallize at about 0.2 and 0.5 mass% Si, respectively, and then increases substantially with silicon.

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Fig. 6 Macrohardness profiles of the samples solidified with and without the AC magnetic field.

Fig. 7 Effects of iron content on the solidification characteristics of Al-Fe-Si alloy predicted from the Gulliver-Scheil model. (Si 0.495 mass%); (a) The relationship between the crystallization temperatures of intermetallic phases and iron content, (b) The relationship between the amount of intermetallic phases and iron content.
content due to the occurrences of eutectic reactions (6) and (7).

In the present study, the major reactions (1) and (2) are clearly evident from the microstructure and DSC measurements. Reaction (3) is supported directly by the higher magnification micrographs in Fig. 5, where the $\alpha$-AlFeSi is growing from Al$_3$Fe. Reactions (4)–(7) are not observed to take place during solidification. The crystallization temperatures of intermetallic phases obtained from DSC measurements are in good agreement with the phase diagram calculations, as shown in Table 2. Two well-defined peaks of two reactions corresponding to the formation of primary aluminum and Al$_3$Fe phases were identified from the DSC curves in Fig. 3. Because the amount of $\alpha$-AlFeSi phase being formed is so small that it would not be observed anyway.

4.2 Distribution of the Al$_3$Fe phase in the AC magnetic field

As shown in Fig. 6, the macrohardness values are the highest at the center and drop sharply at the edge along with the transverse section of the sample. The higher volume fraction of the Al$_3$Fe phase at the center leads to higher hardness, it may be concluded that the sharp drop of the hardness at the edge is attributed to the lower volume fraction of the Al$_3$Fe phase. However, there is no remarkable difference in hardness between the edge and the center when the Al-Fe alloy was solidified in the conventional condition, which indicates the homogeneous distribution of the Al$_3$Fe phase throughout the sample. This result is in good agreement with the structural observations in Fig. 4. The explanations to this phenomenon are discussed as follows.

On the one hand, the rotating melt flow caused by Lorentz force under the AC magnetic field may result in the accumulation of the Al$_3$Fe phase in the center of a whirl. As we all know that, under the effects of the periodic current, the inductor generates a variable magnetic field in the melt, which in turn gives rise to an induced current. Thus, the melt is subject to electromagnetic body forces caused by the interaction of the eddy currents $j$ and the magnetic field $B$. Lorentz force can be solved by Maxwell’s electromagnetic equations as follows:

$$f = j \times B = -\frac{1}{2\mu} \nabla B^2 + \frac{1}{\mu} (B \cdot \nabla)B$$  \hspace{1cm} (8)

where $\mu$ is magnetic permeability. The first term on the right side of eq. (8) is a potential force balanced by a pressure gradient, and the second term on the right side of eq. (8) is a rotational component which results in a forced convection and flow in the melt. However, in our experiment, the magnetic field distribution along the vertical axis of the sample is uniform throughout the sample, i.e.

$$(B \cdot \nabla)B \approx 0$$  \hspace{1cm} (9)

That is to say, the rotational force caused by the Lorentz force is very small and can be negligible here. Therefore, the rotating melt flow inside the melt generated by the rotational force is very weak, which would not lead to the accumulation of the Al$_3$Fe phase.

On the other hand, in the case of electrical conductivity, current density induced by the AC magnetic field varies substantially depending on electrical conductivity of the substances. At the same time, current density determines magnitude of Lorentz force according to eq. (8). That is to say, the higher the electrical conductivity, the larger the Lorentz force. Moreover, the time mean Lorentz force is directed to the center of the sample.\(^{30}\) In the melt, the substance with lower electrical conductivity tends to move toward the edge of the sample while the substance with higher electrical conductivity move toward the center. It is known that the electrical conductivity of the Al melt is much higher than that of the Al$_3$Fe compound.\(^{52}\) Based on this analysis, the Al$_3$Fe phase would be distributed at the edge of the sample, which is contrary to the results obtained in our study. Therefore, there must be some other factor leading to this accumulation.

In view of eq. (9), Lorentz force can be expressed as

$$f = -\frac{1}{2\mu} \nabla B^2 = -\frac{\mu_0}{2} (1 + \chi) \nabla H^2$$  \hspace{1cm} (10)

where $\mu_0$ is vacuum permeability, $\chi$ is magnetic susceptibility and $H$ is applied magnetic intensity. In our solenoid coil, the radial distribution of the magnetic field is not uniform. The smaller the distance from the coil, the higher
the magnetic intensity. The average magnetic gradient from the center to the edge of the coil is 0.3125 T/m, which was determined by CT3-A Tesla Meter at room temperature. Moreover, the magnitude of AC magnetic field inside the melt decreases exponentially from the edge to the center due to the skin effect.\textsuperscript{17} It suggests that the magnetic gradient inside the melt was greatly increased after applying the AC magnetic field. According to eq. (10), Lorentz force, which is in the opposite direction to the magnetic gradient, is directed towards the center of the sample. The difference between Lorentz force acting on the Al matrix \( f_b \) and the Al\textsubscript{1}Fe phase \( f_p \) is written as

\[ \Delta f = f_b - f_p = \frac{\mu_0}{2} (\chi_p - \chi_b) \nabla H^2 \]  

where \( \chi_b \) and \( \chi_p \) are magnetic susceptibility of Al matrix and Al\textsubscript{1}Fe phase, respectively. It can be deduced that the Al\textsubscript{1}Fe phase would be subject to larger Lorentz force if \( \chi_p > \chi_b \). We believe that the magnetic susceptibility of Al\textsubscript{1}Fe phase is higher than that of Al. During the solidification, the Al\textsubscript{1}Fe phase is subject to larger Lorentz force than the Al matrix, which results in the accumulation of the Al\textsubscript{1}Fe phase towards the center.

5. Conclusions

(1) The solidification sequences of this Al-Fe-Si alloy solidified with and without the application of the AC magnetic field were confirmed by the DSC measurements and higher magnification microstructure: primary aluminum dendrite formation; the binary Al-Al\textsubscript{1}Fe eutectic; and \( \alpha \)-AlFeSi peritectic.

(2) Al\textsubscript{1}Fe was the dominant phase in the experimental alloy solidified in two conditions. The amount of the \( \alpha \)-AlFeSi phase with its characteristic dendritic or Chinese script-like morphology was too small to be detected in the DSC curves.

(3) The experimental results obtained in two solidification conditions were consistent with the thermodynamic calculations by the Gulliver-Scheil model. Thermodynamic prediction provided an effective way in studying the effects of alloying elements on the solidification characteristics of the alloy.

(4) The distribution of Al\textsubscript{1}Fe phase was almost homogeneous in the volume of the sample when the alloy was solidified in the conventional condition. With the application of the AC magnetic field, the Al\textsubscript{1}Fe phase was accumulated towards the center of the sample. Macrohardness profiles were in good agreement with the structural observations.

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