Caliber Rolling Process and Mechanical Properties of High Fe-Containing Al–Mg–Si Alloys

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Fe-intermetallic compounds are commonly detrimental for mechanical properties and formability of aluminum alloys. In this study, the refinement of Fe-intermetallic compounds and mechanical properties in high Fe-containing Al–Mg–Si alloys were studied by caliber rolling. The Al-(2.2–2.3)mass%Si-0.9mass%Mg-(1, 1.5 and 2)mass%Fe alloys were severely deformed by the multi-pass caliber rolling at 573 and 723 K. Fe-intermetallic compounds were finely fragmented with the smallest size of around 200 nm. The morphologies of fragmented Fe compounds in the Al matrix depend on the as-cast microstructure and deformation behavior with better distribution in the outer area than in the center area. Good ultimate strength of 345–360 MPa and high elongation of 15–25% were achieved after 95% caliber rolling.

1. Introduction

The recycling process of aluminum scraps commonly contains high amount of impurities, especially Fe content which results in degraded properties compared with the primary aluminum production. The refinement of Fe-intermetallic compounds becomes increasingly important. Generally, the size and shape of Fe-intermetallic compounds can be modified by the control of cooling rate and addition of alloying elements. Recently, the modification of Fe compounds by a mechanical process has been studied. Sato et al. proposed the D-SSC process to refine Fe-intermetallic compounds with good semi-solid microstructures and mechanical properties of the cast Al–Si–Cu–Fe alloy.1) The finely fragmented Fe-intermetallic compound was found as effective particles to refine semi-solid microstructure of the α-Al grain size in the wrought Al–Mg–Si–Fe alloy.2)

However, Fe is intentionally added in the commercial Al alloys to reduced die soldering in the aluminum die casting process.3) Improvement of strength at elevated temperature can be expected by adding of high melting point element into Al alloys.3) Increased amount of Fe contents has also advantage to improve wear behavior.4) Furthermore, the fragmentation and coarsening of the Fe intermetallic compound incorporated with spheroidized Si can improve the wear resistance by a heat treatment.5)

Recently, severe plastic deformation by caliber rolling (CAROL) has been studied for various materials to produce ultrafine grain structures with good toughness improvement and refinement of intermetallic compounds. Mukai et al. observed the refinement of the grain structure of a Mg–Al–Zn alloy by caliber rolling with good improvement of tensile yield stress.6) Severe strain distribution was repeatedly performed during caliber rolling process at the different surface velocities of roller. Inoue et al. investigated the strain distribution in the SM490 steel by a finite element analysis and deformed microstructure observation.7,8) Higher strain accumulation at the corner area of the specimen was found to be higher than the center area.

In this study, high Fe contents between 1–2 mass% (commonly up to 0.1 mass% in the commercial aluminum alloys) were intentionally added to the newly proposed Al–Mg–Si–Fe alloys in order to investigate the negative effects of Fe impurities in the conventional casting and the subsequently improvement by caliber rolling. Severe plastic deformation by caliber rolling was performed to refine the harmful Fe-intermetallic compounds into more favorable size, shape and distribution in the Al-matrix with improved mechanical properties.

2. Experimental Procedure

The chemical compositions of newly proposed Al–Mg–Si–Fe alloys are shown in Table 1. The Si contents are between the commercial wrought Al–Mg–Si 6xxx series alloys and the cast Al–Si–Mg 3xxx series alloys with low amounts of primary Mg2Si and Si phases at 1 mass%Mg and 2.3 mass%Si.9)

Figure 1 shows the schematic illustration of the caliber rolling. Aluminum alloy ingots in the size of 53 × 53 × 250 mm³ were cast by an induction furnace in a steel mold. Then the ingots were homogenized at 783 K for 2h. After that the ingots were sequentially caliber rolled at 573 and 723 K to 70 and 95% area reduction with the cross section area of 28.7 × 28.2 mm² and 11.8 × 11.8 mm², respectively, as shown in Figs. 2 and 3. During caliber rolling, the

Table 1 Chemical compositions of Al–Mg–Si–Fe alloys (mass%).

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Si</th>
<th>Mg</th>
<th>Fe</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>1%Fe</td>
<td>2.2</td>
<td>0.9</td>
<td>0.9</td>
<td>Bal.</td>
</tr>
<tr>
<td>1.5%Fe</td>
<td>2.3</td>
<td>0.9</td>
<td>1.5</td>
<td>Bal.</td>
</tr>
<tr>
<td>2%Fe</td>
<td>2.3</td>
<td>0.9</td>
<td>1.8</td>
<td>Bal.</td>
</tr>
</tbody>
</table>

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specimens were reheated after around 3 passes for 5 min to keep the constant rolling temperatures until the final rolling step. Finally, the T6 treatment was performed at 823 K for 1 h followed by aging at 453 K for 5 h for the precipitation hardening. The characterization was performed with an optical microscope, differential scanning calorimeter (DSC), scanning electron microscope attached with an energy dispersive X-ray spectrometer (SEM/EDS), hardness test and tensile test.

3. Results and Discussions

3.1 DSC analysis

The DSC samples were prepared from the center area of ingots. The DSC heating curves indicate 4 melting reactions as shown in Fig. 4, which correspond to the melting reactions (1)–(4)\(^9\)–\(^{11}\)

\[
\begin{align*}
\text{Al} + \text{Mg}_2\text{Si} & \rightarrow \text{L} \\
\text{Al} + \beta\text{-Al}_3\text{FeSi} & \rightarrow \text{L} \\
\text{Al} + \alpha\text{-Al}_5\text{Fe}_2\text{Si} & \rightarrow \text{L}
\end{align*}
\]

The amounts of intermetallic phases in the Al matrix are estimated based on the area of endothermic peaks. The amounts of Mg\(_2\)Si, Si in reactions (1) and (2) decrease with increasing amount of the Fe content, as a result of reduced Si content which competitively formed by \(\beta\)-Al\(_3\)FeSi and \(\alpha\)-Al\(_5\)Fe\(_2\)Si. On the other hand, the amounts of \(\beta\)-Al\(_3\)FeSi and \(\alpha\)-Al\(_5\)Fe\(_2\)Si in reactions (3) and (4) increase with increasing amount of the Fe content.

3.2 As-cast microstructures

As-cast microstructures of the 1% and 2% Fe alloys are shown in Figs. 5 and 6. The type and morphology of the Fe-intermetallic compounds strongly depend on alloy compositions and solidification conditions of the as-cast ingot. Segregation of the Fe-rich regions (regions with high number density of Fe-intermetallic compounds) in the center resulted in various types and shapes of Fe-intermetallic compounds. On the other hand, high cooling rate in the outer resulted in the fine Fe-intermetallic compounds. The plate-like shape of \(\beta\)-Al\(_3\)FeSi was found as more harmful than \(\alpha\)-Al\(_5\)Fe\(_2\)Si because of its size, sharp edge and weak interface with the Al matrix which can initiate cracking at the interface between the Fe-intermetallic compound and the Al matrix.\(^{12}\)

Fine plate-like Fe-intermetallic compounds at the grain boundary of the \(\alpha\)-Al dendrite were found in the outer area with high cooling rate as shown in Figs. 5(a), 5(b) and 6(a) which correspond to the reaction (3). Fe-rich regions were found in the center area as shown in Figs. 5(d) and 5(f). Primary polyhedral \(\alpha\)-Al\(_5\)Fe\(_2\)Si was found in the center area of the 1.5% Fe and 2% Fe alloys as shown in Figs. 5(f) and 6(d), which corresponds to the ejection of Fe concentration into the \(\alpha\)-Al\(_5\)Fe\(_2\)Si region.\(^{11}\)

3.3 Caliber rolling

3.3.1 Caliber-rolled microstructures

Caliber-rolled microstructures are shown in Figs. 7 and 8. Inhomogeneous distribution of Fe compounds was found
especially in the center after 95% caliber rolling. Inoue et al.\textsuperscript{7} found that high strain accumulation was generated by high deformation ratios of caliber rolling, especially in the outer area of the specimen. Good fragmentation and distribution of Fe-intermetallic compounds were found in the outer area as shown in Figs. 7(a), 7(b), 8(a) and 8(b). Higher density of fragmented Fe-intermetallic particles was found in the 2\%Fe alloy. On the other hand, less fragmentation and distribution of Fe-intermetallic compounds were found in the center area as shown in dotted circles in Figs. 7(c), 7(d), 8(c) and 8(d), because of less deformation compared with the outer area and high segregation of the Fe-rich regions in the center area of the as-cast ingot as shown in Figs. 5(e) and 5(f). Especially the Chinese script $\alpha$-Al$_8$Fe$_2$Si tends to locate locally deformed in an elongated distribution after caliber rolling as shown in Figs. 7(d) and 8(d). Large polyhedral $\alpha$-Al$_8$Fe$_2$Si compounds are remained in the center area as shown in Fig. 8(d) in the 1.5 and 2\%Fe alloys.

![Fig. 5 Microstructures of Fe-intermetallic compounds: the 1\%Fe as-cast alloy in outer area (a), intermediate area (c) and center area (e) and the 2\%Fe as-cast alloy in outer area (b), intermediate area (d) and center area (f).](image-url)
The Fe-intermetallic compounds became finely fragmented as shown in the SEM image in Fig. 9. The smallest size of the fragmented Fe-intermetallic particles was around 200 nm in the magnified area of Fig. 9(a) as shown in Fig. 9(b). Severe plastic deformation of binary Al–Fe alloys was investigated by the high-pressure torsion (HPT).\cite{13-15} It was found that such severe deformation can lead to the solid solution extension of Fe in the Al matrix, which was confirmed by the lattice constant of Al and artificial age hardening. Inoue \textit{et al.}\cite{8} evaluated by a finite element analysis that the equivalent strain $\varepsilon_{eq}$ introduced by 94% caliber rolling was about 7. On the other hand, considerably higher equivalent strain $\varepsilon_{eq}$ of 633 (calculated from the true logarithmic strain, $\varepsilon_{log} = 7$) by Senkove \textit{et al.}\cite{13,14} and up to 2500 by Cubero-Sesin \textit{et al.}\cite{15} were performed by HPT. By comparison of these equivalent strain $\varepsilon_{eq}$ accordingly, the
applied strain by 95% caliber rolling might not be sufficient to induce solid solution of the Fe-intermetallic compounds in the Al matrix. However, the details of solid solubility of Fe as well as Fe-intermetallic compounds have not been experimentally confirmed in this study.

The rolling temperatures of the high Fe-containing Al alloys were carefully considered because the Fe-intermetallic compounds are detrimental to the formability, especially in the rolling process. Caliber rolling at 573 K was prone to initiate severe cracks in the center area of the specimen along the rolling direction, while the cracks could be avoided by higher temperature rolling at 723 K. The morphologies of fragmented Fe compounds were found to be similar at both rolling temperatures with slightly better distribution of Fe compounds at 723 K as shown in Fig. 10. The perpendicular cross section micrograph to the rolling direction of the 95% caliber-rolled specimen at 573 K shows high amount of Fe-rich regions (as indicated with dotted circles) in the center area of the specimen along the rolling direction, while the cracks could be avoided by higher temperature rolling at 723 K. The morphologies of fragmented Fe compounds were found to be similar at both rolling temperatures with slightly better distribution of Fe compounds at 723 K as shown in Fig. 10. The perpendicular cross section micrograph to the rolling direction of the 95% caliber-rolled specimen at 573 K shows high amount of Fe-rich regions (as indicated with dotted circles) in the center area of the specimen in Fig. 10(a), whereas better distribution of the Fe-rich regions was found in the caliber-rolled specimen at 723 K in Fig. 10(b). The parallel cross section to the rolling direction of Fe-rich regions is shown in the elongated band in Fig. 7(c). The flow of the Al matrix and the second phase particles such as Fe-intermetallic compounds become easier with increasing temperature. Therefore, the fragmented Fe-rich regions became more sheared, embedded and distributed in the softer Al matrix at higher temperature.

3.3.2 EDS analysis

The chemical compositions of fragmented Fe-intermetallic compounds in Fig. 11 were quantitatively confirmed by the EDS analysis based on the Fe : Si atomic ratio\(^ {11}\) as listed in Table 2. Large polyhedral Fe-intermetallic compounds in Fig. 11(a) were indicated as $\alpha$-Al\(_8\)Fe\(_2\)Si with Fe : Si ratio of 1.72 : 1 and 1.62 : 1 which are close to 2 : 1 of $\alpha$-Al\(_8\)Fe\(_2\)Si. The large primary $\alpha$-Al\(_8\)Fe\(_2\)Si is located in the center area of the ingot and difficult to be fragmented as shown in Fig. 8(d). The small Fe-intermetallic compounds in Fig. 11(b) were indicated as fragmented $\beta$-Al\(_5\)FeSi with Fe : Si ratio of 0.97 : 1 and 0.81 : 1 which are close to 1 : 1 of $\beta$-Al\(_5\)FeSi. However, the smaller fragmented Fe-intermetallic compounds could not be properly analyzed due to the thin shape of Fe compounds which leads to include high matrix signal.
3.3.3 Fracture surfaces

Various fractures of the tensile specimen were observed as shown in Fig. 12, which demonstrates the fracture characteristics of the distribution of fragmented Fe-intermetallic compounds. Inhomogeneous distribution of the fragmented Fe compounds is shown in Fig. 12(a) with fractures of the Fe-rich region, low number density of Fe compounds and good distribution of Fe compounds. Figure 12(b) shows the mixed region of low density and high density of Fe compounds. The low number density of Fe region shows dimple fracture of the Al matrix with distributed Fe compounds in Figs. 12(b) and 12(e). The Fe-rich region shows short interparticle spacing of Fe compounds surrounded by the Al matrix in Fig. 12(d). Good distribution of Fe compounds in Fig. 12(c) shows probably the best combination of fragmented Fe compounds in the Al matrix, which can combine good ductility of the Al matrix and refined strengthening Fe compounds. Small amount of large polyhedral primary Fe compounds have cleavage fracture crossing its particle as shown in Fig. 12(f).

3.3.4 Mechanical properties

After caliber rolling, all specimens were performed precipitation hardening by the T6 treatment. Ultimate strength (UTS) and elongation of the 70 and 95% caliber-rolled specimens and the as-cast 1%Fe alloy are summarized in Fig. 13. The 70% caliber-rolled specimens show UTS between 338–351 MPa with elongation between 7.8–12.0%. Higher deformation by 95% caliber rolling shows good UTS between 345–360 MPa with elongation between 14.6–24.8%. UTS and elongation of both 70 and 95% caliber-rolled specimens are clearly higher than that of the 1%Fe as-cast specimen (269 MPa). The elongation of 15–25% in the 95% caliber-rolled specimen is greatly improved compared with only 1.5% elongation in the 1%Fe as-cast specimen. However, no significant difference in the mechanical properties was found between 573 and 723 K caliber rolling. The negative effect of large Fe intermetallic compounds was effectively modified by caliber rolling. Brittle fracture surfaces between the large Fe-intermetallic compounds and the Al matrix of the 1%Fe as-cast specimen are shown in...
The crack initiation and propagation occurred along the interface between the Fe intermetallic compounds and the Al matrix as shown in Fig. 14(a). Brittle cleavage fractures were observed along the large plate-like \( \beta \)-Al\(_5\)FeSi as shown in Fig. 14(b).

With the refinement of Fe compounds by caliber rolling, Fe intermetallic compounds becomes finely fragmented and distributed in the Al matrix. Figure 12(c) shows finely fragmented Fe particles surrounded by the fine dimple fracture of the Al matrix. The cracking propagation is decelerated by passing the Al matrix and the refined fragmented Fe compounds, consequently the strength and elongation are improved. Nevertheless, more investigation of the strengthening mechanism by distribution of the fine Fe-intermetallic phase in the Al matrix is still required, as various cracking mode were observed in Fig. 12.

In comparison with some commercial Al alloys with the T6 treatment, the Al–Si–Mg 356.0 alloy has UTS of 262 MPa and elongation of 5%\(^{16}\) and the Al–Mg–Si 6082 alloy has UTS of 315 MPa and elongation of 12%.\(^{17}\) Generally, the Fe contents are minimized at 0.1 mass% in these commercial alloys. The 70% caliber-rolled specimens are comparable or superior of those commercial Al alloys in both UTS and elongation. The 95% caliber-rolled specimens show great improvement in elongation significantly compared with those commercial alloys. Note that the caliber-rolled specimens contain much higher amount of Fe impurities than the commercial Al alloys.

The strengthening mechanism from fine second-phase particles such as fragmented Fe-intermetallic compounds in the Al matrix depends on the distribution of particles, shape, particle size and interparticle spacing.\(^{18}\) In the case of the distribution of fragmented Fe compounds in the Al matrix, Fe-intermetallic compounds are considered as the hard and brittle phase in the ductile \( \alpha \)-Al matrix. The stress–strain curves in Fig. 15 represent the typical tensile properties of the 70 and 95% caliber-rolled specimen after the T6 treatment. These graphs show stress–strain curves of the typical metal matrix composites with 3 stages; I, II and III.\(^{19,20}\) In the stage I both the Al matrix and fragmented Fe-intermetallic compounds undergo elastic deformation. In the stage II the Al matrix undergo plastic deformation while the fragmented Fe-intermetallic compound retains plastic deformation. Finally, in the stage III both Al matrix and fragmented Fe-intermetallic compounds undergo plastic deformation with parabolic curve until fracture. During caliber rolling, the specimen was directionally deformed. The plate-like \( \beta \)-Al\(_5\)FeSi as well as Chinese-script \( \alpha \)-Al\(_3\)Fe\(_2\)Si, which also consist of thin plate in the network shape,\(^{12}\) were finely fragmented and preferentially aligned on the rolling direction as shown in Figs. 7(b), 7(d), 8(b) and 8(d), which can enhance the mechanical properties along the rolling direction.

The refinement of distribution of Fe-intermetallic compounds by caliber rolling is an effective process to modify the harmful Fe intermetallic compounds into strengthening phase by refinement and distribution.
4. Conclusions

The modification of high Fe-impurity containing Al–Mg–Si alloys by caliber rolling was investigated. The results are summarized as follows.

(1) The refinement of harmful Fe-intermetallic compounds by multi-pass caliber rolling shows good modification of harmful Fe-intermetallic compounds into more favorable size, shape and distribution of Fe-intermetallic particle.

(2) Significant improvement of ultimate strength (345–360 MPa) and elongation (15–25%) was achieved by 95% caliber rolling in comparison with the unmodified as-cast specimen.

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