Effects of Grain Boundary Characteristics on Secondary Recrystallization Textures in Fe–Si Alloy

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To understand the factors that determine the secondary recrystallization textures of Fe–Si alloy, the change in the secondary recrystallization textures of Fe–Si sheets was investigated by increasing the cold-rolled reduction rate (CR) from 70 up to 95%.

The secondary recrystallization textures in the CRs=90–95% samples accumulated in specific orientations; the main components of the secondary recrystallization were \{110\}<001> in the CR=90 and 93% samples, and \{110\}<001> with \{110\}<112> in the CR=95% sample. The experimental results were reproduced by calculations based on the idea that the secondary recrystallization texture is mainly determined by the frequency of the specific-orientation-grain-boundaries, for example, coincidence-site-lattice grain boundaries. In contrast, in the CR=70% sample, the secondary recrystallization texture was not accumulated but dispersed from \{110\}<001> to \{110\}<225> and was inaccurately reproduced by the above calculations.

The study concludes that the secondary recrystallization textures in the CRs=90–95% samples are mainly determined by the grain boundary effect. It is also concluded that the secondary recrystallization textures of the CR=70% sample are determined by both grain boundary effect and nucleus effect. The difference in the mechanisms originates from the changes in the frequency of the specific-orientation-grain-boundaries in the matrix and of the nuclei at the surface in the primary recrystallization textures of various CRs.

KEY WORDS: recrystallization; texture; grain boundary; silicon steel; Goss; \{110\}<001>.

1. Introduction

Secondary recrystallization can be obtained by re-annealing recrystallized materials, in other words, the primary recrystallization materials. The main component of the secondary recrystallization texture in Fe–Si sheets is well known as the \{110\}<001> orientation.1–4) In other words, the Goss orientation. Because the axis of easy magnetization of iron is \<001>, the secondary recrystallized sheets of Fe–Si are used in the core of transformers using the magnetic flux. To improve the magnetic properties of the Fe–Si sheets by controlling the secondary recrystallization textures, it is important to understand the orientation selectivity mechanism during secondary recrystallization of Fe–Si sheets.

Thus, many studies5–26) have been conducted to clarify the factors which determine the secondary recrystallization textures of Fe–Si sheets. Among them, two conventional hypotheses20–25) explaining the orientation selectivity during secondary recrystallization have been proposed. One is the coincidence-site-lattice (CSL) grain boundaries model20–22) (CSL model), and the other is the high energy (HE) grain boundaries model23,24) (HE model). Although these models are similar in the idea that the grain boundary characteristics play an important role in determining the secondary recrystallization textures, they are different in some aspects as explained below.

In the CSL model, the \Sigma 9-CSL grains, which are obtained by rotating the Goss grains by 38.9° around the \<110> axes, are preferentially consumed by the Goss grains. Assuming that the grain boundary energies between the Goss grains and the CSL grains are lower than those between the Goss grains and the other-orientation grains, the Goss grains...
experience a lower pinning force due to precipitates (inhibitors). Consequently, the Goss grains preferentially consume the matrix.\textsuperscript{20–22)}

In the HE model proposed by Hayakawa and Szpunar,\textsuperscript{23,24)} the HE grains, which have misorientation angles of 20–45° from the Goss orientation, are preferentially consumed by the Goss grains. They suggest that HE grain boundaries have a relatively small pinning force by the inhibitor due to faster dissolution by the rapid grain boundary diffusion.

To evaluate the validity of the models, Arai \textit{et al.}\textsuperscript{8,12) investigated the effect of the cold-rolled reduction rate (CR) on the secondary recrystallization textures. They suggested that the main components in the secondary recrystallization textures in Fe–Si steel sheets, where the final sheet thickness effects.

To clarify the factors determining the secondary recrystallization textures in detail, the relation between the primary recrystallization textures and the secondary recrystallization textures of the Fe–Si by changing the CR. From this work, the main components in the secondary recrystallization textures of Fe–Si sheets cold-rolled at 88% and 95% were \{110\} <001> and \{110\} <112>, respectively. Additionally, Imamura \textit{et al.}\textsuperscript{12) reported the geometric relationship between the primary recrystallization textures and the secondary recrystallization textures of the Fe–Si by changing the CR. From this work, the main components in the secondary recrystallization textures of Fe–Si sheets cold-rolled at 92.6% and 97.2% were \{110\} <001> and \{110\} <112>, respectively. They claimed that the calculation based on the HE model reproduces their experiments in the CR = 97.2% sample, but the calculation based on the CSL model was inconsistent with their experiment. However, in their work,\textsuperscript{8,12) the final sheet thickness was different with CR. Because the orientation selectivity of the secondary recrystallization is also affected by the final sheet thickness,\textsuperscript{8,10,25–27) their results\textsuperscript{8,12) are expected to be affected by the final sheet thickness effects.

To clarify the factors determining the secondary recrystallization textures in detail, the relation between the primary recrystallization textures and the secondary recrystallization textures in Fe–Si steel sheets, where the final sheet thickness is the same, at various CRs was studied in this paper.

2. Materials and Methods

The materials used in this work were hot-rolled Fe-3mass%Si sheets. The chemical composition followed the previous work by Arai \textit{et al.}\textsuperscript{8) that is, Fe, 3mass% Si, 0.05mass% C, 0.1mass% Mn, and 0.03mass% Al. The thickness of the hot-rolled Fe–Si sheets were 2.3, 3.2, and 4.5 mm. The hot-rolled Fe–Si sheets were annealed at 1 373 K in a nitrogen gas atmosphere. The specimens were cold-rolled to the same thickness of 0.23 mm. The CRs were 90%, 93%, and 95% for the hot-rolled bands with the thicknesses of 2.3, 3.2, and 4.5 mm, respectively. In this work, samples cold-rolled at 70% were also prepared as follows: the 2.3 mm thick annealed hot-rolled bands were cold-rolled to 0.80 mm and then annealed at 1 173 K for 120 s. The 0.80 mm thick annealed samples were cold-re-rolled to 0.23 mm. Table 1 lists the changes in the gauge thickness of the samples in each experimental process.

Then, the primary recrystallization sheets were prepared, that is, all the cold-rolled sheets were annealed at approximately 1 100 K for 100 s in a mixed nitrogen and hydrogen atmosphere followed by a nitrogen injection. Next, to enable secondary recrystallization, the primary recrystallization sheets were re-annealed from room temperature up to 1 473 K at a heating rate of approximately 15 K/h in the mixed nitrogen and hydrogen atmosphere.

The textures of the secondary recrystallization samples were measured by the back-reflection Laue diffraction method, with measurement areas of approximately 60 × 100 mm for each sample.

The primary recrystallization textures at the surface (t = 1/10) and the center (t = 1/2) positions of the samples were measured by an electron back scattered diffraction pattern (EBSD) method, where the sample dimensions were 10 × 15 mm. According to earlier studies,\textsuperscript{8–10) the analysis on the matrix texture, in other words, the texture at the center position of the sample, is suitable for identifying the grains preferentially consumed by the nuclei in the secondary recrystallization. In contrast, the analysis of the surface texture is a good indicator of the frequency and orientations of the secondary recrystallization nuclei, which exist mainly in the surface positions of the sample.

Mechanical polishing and electrolytic polishing were adopted for thickness reduction and surface polishing, respectively, to obtain the Kikuchi patterns. The measurement areas were 20 mm\textsuperscript{2} with 4 μm steps and 40 mm\textsuperscript{2} with 80 μm steps for the surface (t = 1/10) positions and the center (t = 1/2) positions, respectively.

In this paper, the experimentally obtained secondary recrystallization textures are compared with the calculated secondary recrystallization textures based on the CSL and the HE models. The calculated secondary recrystallization textures were computed from the primary recrystallization textures obtained by the EBSD measurements as described below:

In the case of the CSL model,\textsuperscript{20–22) the experimental primary recrystallization textures were rotated by 38.9° around twelve kinds of the <110> axes. All the rotated textures around the <110> axes were merged and plotted in the orientation distribution function (ODF) maps. According to the CSL model, the Goss nuclei selectively consume the Σ9 grain boundaries, and the Σ9 grain boundaries are defined by the 38.9° rotation around the twelve kinds of <110> axes.

In the case of the HE model,\textsuperscript{23,24) all the reference angle combinations, in other words, from (φ1, Φ, φ2) = (0, 0, 0) to (φ1, Φ, φ2) = (2π, π, 2π), were compared with the

<table>
<thead>
<tr>
<th>CR</th>
<th>Hot-rolled sheet</th>
<th>Annealed hot-rolled sheet</th>
<th>Cold-rolled sheet</th>
<th>Re-annealed hot-rolled sheet</th>
<th>Cold-re-rolled sheet</th>
</tr>
</thead>
<tbody>
<tr>
<td>70%</td>
<td>2.3 mm → 2.3 mm → 0.80 mm → 0.80 mm → 0.23 mm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>90%</td>
<td>2.3 mm → 2.3 mm → 0.23 mm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>93%</td>
<td>3.2 mm → 3.2 mm → 0.23 mm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>95%</td>
<td>4.5 mm → 4.5 mm → 0.23 mm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Table 1. Changes in the sample gauge thickness of from the hot-rolled sheets to the cold-rolled sheets.*
experimentally obtained primary recrystallization textures. The number of HE grains, which have misorientation angles of 20–45°, in each reference angle of (φ₁, Φ, φ₂) were counted. Each reference angle of (φ₁, Φ, φ₂) versus the number of HE grains were plotted in the ODF map.

3. Experimental Results

Figure 1 depicts the secondary recrystallization structures. It can be seen that the grain size of the CR=70% sample was smaller than the other CR samples.

Figure 2 depicts the secondary recrystallization texture ODF maps. In the CR=95% sample, the main component was split into the {110} < 001 > to {110} < 112 > orientations. It was noted that the components in the CR=95% sample were split but the deviations from each orientation were small, in other words, “sharp”. In the CR=90 and 93% samples, the main component was sharp {110} < 001 > orientation. In the secondary recrystallization textures of the CR=70% sample, the orientations were continuously dispersed from {110} < 001 > to {110} < 225 >.

The changes in the primary recrystallization textures with CR are shown because the changes in the secondary recrystallization textures should originate from the primary recrystallization textures. Figure 3 depicts the ODF maps of the primary recrystallization textures which were experimentally obtained by the EBSD measurements of the center (t=1/2) parts. In Fig. 3(a), it can be seen that the {111} < 112 > and {411} < 148 > components were dominant in the CRs=90–95% samples, which are known to be the Σ9-CSL grain boundaries for the Goss orientation.8–10 From the Figs. 3(b) and 3(c), {11 5 4} < 5 19 10 > and {411} < 2 11 19 > components also increased with the CR, which are the Σ9-CSL grain boundaries for {110} < 112 >. While the primary recrystallization textures of the CRs=90–95% were sharp, that of the CR=70% was dispersed.

Figure 4 depicts the predicted secondary recrystallization textures based on the CSL and HE models. Both calculated secondary recrystallization textures are in good agreement with the experimental ones in the CRs=90–95% samples, as listed in Table 2. However, both models could not accurately reproduce the experimental results for the CR=70% sample, that is, the CSL and HE models predicted the secondary recrystallization to be {110} < 225 > and {110} < 114 >, respectively, but the dispersion from {110} < 001 > to {110} < 225 > was not accurately reproduced.

Figure 5 depicts the abundance ratios of the {110} < uvw > nuclei frequency for the various CRs, which were observed from the primary recrystallization textures at the surface (t=1/10) parts of the samples. Large amounts of nuclei exist only in the CR=70% sample. Alternatively, the number of nuclei was very small in the cold-rolled samples at more than 90% CR.

4. Discussion

As in previous studies,22,26,28–30 the reason that the cal-
Calculations from this study were in good agreement with the texture of the CRs = 90–95% samples is that even if the frequency of nuclei is very small, secondary recrystallization occurs via the grain boundary effect in the following conditions:

i) That a large frequency of specific-orientation-grain-boundaries, for example, coincidence-site-lattice grain boundaries, is available in the primary recrystallization in the matrix.

ii) That the precipitation behavior and thermal stability of the inhibitors are properly controlled.

In this type of secondary recrystallization, the structures feature sharp textures and a large grain size. The sharp texture originates from the specific-orientation-grain-boundaries which are preferentially consumed only by specific-orientation-nuclei. For example, the \{111\} <112> and \{411\} <148> orientations have specific-orientation-grain-boundaries for the Goss orientation. In the secondary recrystallization, the Goss orientation preferentially consumes those grain boundaries (CSL grain boundaries) according to the CSL model. Consequently, the orientation dispersion of the secondary recrystallization texture becomes sharp.

The latter feature (large grain size) is acceptable in the point that the specific-orientation nuclei only grow into the secondary recrystallization grains by consuming large volumes of specific-orientation-grains in the matrix. The secondary recrystallization textures in the CRs = 90–95% samples are likely to be mainly determined by the grain boundary effect, because the secondary recrystallization

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**Fig. 3.** Experimentally obtained primary recrystallization texture (t=1/2) changes in the various CRs. (a) \(\varphi_2=45^\circ\), (b) \(\varphi_2=70^\circ\), and (c) \(\varphi_2=75^\circ\). (Online version in color).
structures of those samples satisfy the above two features, in other words, sharp textures and large grains. Because the calculation used in the study was based on the idea that the secondary recrystallization texture is determined by the geometric relationship between the matrix and nuclei (grain boundary effect), the calculations were in good agreement with the experiments in the CRs $= 90–95\%$ samples.

According to the CSL model, we discuss the secondary recrystallization textures with CR. In the case of CR $= 90$ and 93%, the Goss orientation was sharp. This comes from the large frequencies of $\{111\} < 112 >$ and $\{411\} < 211 >$ grain boundaries ($\Sigma9$-CSL grain boundaries for ideal Goss). In the case of CR $= 95\%$, the secondary recrystallization texture was split into the Goss and $\{110\} < 112 >$ orientations. The $\{110\} < 112 >$ orientations in the case of CR $= 95\%$ comes from the large frequencies of $\{115 4\} < 5 19 10 >$ and $\{411\} < 2 11 19 >$ grain boundaries, which are the $\Sigma9$-CSL grain boundaries for $\{110\} < 112 >$.

However we note that the grain boundary effect is weakened by the nucleus effect in case that there are many nuclei in the primary recrystallization structures. For example, Harase et al. have suggested that the probability of the secondary recrystallization is proportional to the product of the frequencies of CSL grain boundaries (the grain boundary effect) and those of nuclei (the nucleus effect). We consider that the secondary recrystallization structure via the nucleus effect features relatively small grain sizes, because many nuclei grow into secondary recrystallization grains with crowding together.

Considering that this feature was observed in the CR $= 70\%$ sample in Fig. 1, the disagreement between the calculations and the experimental results in the CR $= 70\%$

<table>
<thead>
<tr>
<th>CR (%)</th>
<th>70</th>
<th>90</th>
<th>93</th>
<th>95</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varphi1$ (0.0-90.0 deg.)</td>
<td>$\varphi1$ (0.0-90.0 deg.)</td>
<td>$\varphi1$ (0.0-90.0 deg.)</td>
<td>$\varphi1$ (0.0-90.0 deg.)</td>
<td></td>
</tr>
<tr>
<td>CSL Frequency of 232:CSL grain boundaries (%)</td>
<td>0</td>
<td>30</td>
<td>60</td>
<td>90</td>
</tr>
<tr>
<td>Max $= 1.68$</td>
<td>1.75</td>
<td>1.65</td>
<td>1.55</td>
<td>1.45</td>
</tr>
<tr>
<td>HE Frequency of HE grain boundaries (%)</td>
<td>0</td>
<td>30</td>
<td>60</td>
<td>90</td>
</tr>
<tr>
<td>Max $= 44.27$</td>
<td>43.00</td>
<td>40.00</td>
<td>37.00</td>
<td>34.00</td>
</tr>
</tbody>
</table>

Fig. 4. Calculated secondary recrystallization textures for the various CRs, based on the CSL and HE models. (Online version in color).

Table 2. Comparison of the experimentally obtained secondary recrystallization textures and the calculated secondary recrystallization textures in the various CRs.

<table>
<thead>
<tr>
<th>CR (%)</th>
<th>70</th>
<th>90</th>
<th>93</th>
<th>95</th>
</tr>
</thead>
<tbody>
<tr>
<td>Expt.</td>
<td>${110} &lt; 001 &gt;$</td>
<td>${110} &lt; 225 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
</tr>
<tr>
<td>Calc.(CSL)</td>
<td>${110} &lt; 225 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
</tr>
<tr>
<td>Calc.(HE)</td>
<td>${110} &lt; 114 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
<td>${110} &lt; 001 &gt;$</td>
</tr>
</tbody>
</table>

Table 2. Comparison of the experimentally obtained secondary recrystallization textures and the calculated secondary recrystallization textures in the various CRs.
sample originates from the combination of the grain boundary effect and the nucleus effect, because the nucleus effect is not taken into account in the calculations.

The study verified the suggestion that the grain boundary effect and the nucleus effect coexist in the CR=70% sample. Figure 6 depicts the relationship between the grain sizes of the secondary recrystallization and its orientations in the CR=70% sample. An example of the relation between the structure and the crystal orientation is also shown in Fig. 7. Because the average grain size of the secondary recrystallization is greater than 10–20 mm in general,8,9) in this study only grains greater than 15 mm were analyzed.

It is visible in Fig. 6 that the average grain size increased with the orientation deviation from \{110\}<001> to \{110\}<225>. For example, the grain sizes of \{110\}<114> and \{110\}<225> were relatively large. From this result, it is considered that the grain boundary effect increases with the orientation deviation from \{110\}<001> to \{110\}<225>. Indeed, the calculations in this study predicted the secondary recrystallization textures to be \{110\}<114> or \{110\}<225> in the CR=70% sample. In contrast, it is considered that the nucleus effect increases with the orientation deviation from \{110\}<225> to \{110\}<001>. This is further supported by the increase in the frequencies of nuclei with the orientation deviation from \{110\}<225> to \{110\}<001> in Fig. 5.

To summarize the above discussion, the secondary recrystallization textures in the CRs=90–95% samples are mainly determined by the grain boundary effect. The sec-
ondary recrystallization textures of the CR = 70% sample are determined by a mixture of the grain boundary effect and the nucleus effect. The grain boundary effect mainly originates from the high frequency of specific-orientation-grain-boundaries in the matrix, for example, CSL grain boundaries according to the CSL model. In contrast, the nucleus effect mainly originates from the high frequency of nuclei in the surface of the samples.

From this study, it is evident that the grain boundary effect is important in promoting the orientation selectivity and, consequently, the sharpness of the secondary recrystallization textures as in the case of CRs = 90–95%. Because the sharp texture of the secondary recrystallization of Fe–Si results in high permeability, the authors suggest that utilizing the grain boundary effect is key to improving transformer efficiency. The authors also highlight the importance and validity of the CSL model to precisely predict the sharp textures of the secondary recrystallization.

5. Conclusions

The effects of CR on the secondary recrystallization textures and the primary recrystallization textures of Fe–Si sheets were experimentally investigated by back-reflection Laue diffraction and EBSD measurements, respectively. The secondary recrystallization textures changed with differing CR. In the CRs = 90 and 93% samples, the {110} < 001 > orientation was the main component of the secondary recrystallization textures. In the CR = 95% sample, the {110} < 001 > with {110} < 112 > orientation was the main component of the secondary recrystallization textures. In the CR = 70% sample, the secondary recrystallization textures were continuously dispersed from {110} < 001 > to {110} < 225 >.

The calculations based on the CSL and the HE models were in good agreement with the experimentally obtained secondary recrystallization textures in the CRs = 90–95% samples. However, in the CR = 70% sample, the calculations could not accurately reproduce the experimental observations. The secondary recrystallization textures in the CRs = 90–95% samples are likely to be mainly determined by the grain boundary effect in the presence of a high frequency of the specific-orientation-grain-boundaries in the matrix, for example, CSL grain boundaries according to CSL model. In contrast, the secondary recrystallization textures in the CR = 70% samples are likely to be determined by not only the grain boundary effect, but also by the nucleus effect in the presence of a high frequency of nuclei at the surface.

From this study, the authors suggest that the grain boundary effect is important in promoting the orientation selectivity and, consequently, the sharpness of the secondary recrystallization texture as in the case of CRs = 90–95%. Considering that the sharp texture results in the high permeability of Fe–Si, the authors conclude that utilizing the grain boundary effect is key to improving transformer efficiency. The authors also highlight the importance and validity of the CSL model to precisely predict the sharp textures of the secondary recrystallization.

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