Grain Boundary Characteristics in Grain Oriented Silicon Steel

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During secondary recrystallization of grain oriented silicon steel, isolated grains are often observed in secondary-recrystallized Goss grains (matrix). In an extraction experiment, the crystallographic relations between each isolated grain and the matrix were investigated. Although the $\Sigma 9$ grain boundary against Goss orientation is the most frequent in the primary texture, it could not be observed in the relation, but specific grains, which have $\Sigma 3$ or a high angle misorientation relation, were often observed. As the extraction temperature increased, the frequency of $\Sigma 3$ decreased and that of GGB (general grain boundary) increased. It was revealed that the $\Sigma 9$ boundary was mobile, $\Sigma 3$ was less mobile and GGB was the least mobile. This does not contradict the CSL model but supports it. However, it was not clarified whether this result is caused by grain boundary energy or mobility.

KEY WORDS: electrical steel; silicon steel; grain boundary; recrystallization texture.

1. Introduction

Grain Oriented Silicon Steel (GO) has been mainly used as the core material in transformers, and it is the only product manufactured in the steel industry that applies the secondary-recrystallization phenomenon. The most successful texture control has been achieved on the industrial scale.1) Its magnetic properties, low core loss and high permeability as the core material in transformers, and it is the only product manufactured in the steel industry that applies the secondary-recrystallization phenomenon. The most successful texture control has been achieved on the industrial scale.1) Its magnetic properties, low core loss and high permeability especially the $\Sigma 9$ boundary, which play a significant role by their lower grain boundary energy and/or higher mobility. In the latter model, the grain boundaries have high energy and play a significant role.

Incidentally, isolated (island) grains in secondary-recrystallized Goss grains (matrix) during the secondary-recrystallization annealing have been frequently observed. The reason why these isolated grains can survive up to higher temperature should contain some metallurgical inevitability. K. T. Lee et al.16) reported the change in texture of CGO (Conventional grain oriented silicon steel: two stages of the cold rolling method) with extraction annealing by ODF (Crystallite Orientation Distribution Function) analysis, and that the $\Sigma 5$ boundary might play a significant role. Furthermore, we17) have already reported based on the CSL model that the $\Sigma 9$ boundary might be the fastest boundary of all the boundaries and the $\Sigma 5$ boundary might be the second fastest boundary possibly caused by its lower grain boundary energy.

This study was conducted to indirectly verify the CSL model from the perspective of grain boundary characteristics by measuring the crystallographic relations between isolated grains and Goss grains (matrix) in the extraction experiment during secondary-recrystallization annealing of high permeability GO material.

2. Experimental Procedure

Table 1 shows the chemical composition of the steel used. The specimens, which were decarburized and primary-recrystallized, were prepared from commercial production.16,17) The main inhibitors are finely dispersed AlN and MnS. It was cold-rolled to a thickness of 0.285 mm with a reduction of 87.6%. The annealing separator, which mainly consists of MgO, was coated on the specimen surfaces.

2.1. Primary Recrystallization Texture

The primary recrystallization textures were measured by the X-ray diffraction method before the extraction experiment. They were evaluated by the complete \{100\} pole figures measured at 9 depths from one surface to another through the entire thickness. The thickness of the speci-
mens was around 60 μm. Nine specimens for one series were taken from each 10% position: 10.5, 21.1, 31.6, 42.1, 49.8, 57.9, 68.4, 78.9 and 89.5%. Three series were measured in order to improve the precision. The data were averaged at each position and turned down at the central position, i.e., the datum at the central position was the average of 3 data and the data at other positions were the average of 6 data. These data were three-dimensionally analyzed by the Vector method.20) The intensity of any coincidence orientation,9,21) $S_i$, for a recognized nucleus orientation is expressed as $I_{cS_i}$, which means the percent for total intensity. In this study, it was Goss orientation ({110}/[001], {110}/[111]) and $I_{cS_i}$ ($i = 1–29$), was calculated.

2.2. Extraction Experiment

The specimens coated with MgO were extracted during secondary-recrystallization annealing, which was performed in a 25% N$_2$ and 75% H$_2$ atmosphere from room temperature to 1 373 K with a heating rate of 15 K/h. They were extracted at 25 K intervals from 1 198 to 1 373 K.

2.3. Misorientation between an Isolated Grain and Matrix

Cross sectional microstructures were observed for the transverse direction. Figure 1 shows the cross sectional microstructure of specimens extracted, being extracted at 1 248 K (a) and at 1 273 K (b). (c) shows the colonies of isolated grains. Isolated grains can be observed in large secondary-recrystallized grains (matrix grain). The crystallographic relations between one isolated grain and one surrounding secondary-recrystallized matrix grain were measured with EBSD on the cross section. Although a colony of isolated grains could sometimes be observed, in order to simplify and purify the relation, one isolated grain at each measurement was noted. However, even if two-dimensional observation shows the isolated grain (non colony), it does not always guarantee that the grain is a perfectly three-dimensionally isolated grain.

The specimens of the 8 extraction temperatures every 25 K from 1 198 to 1 373 K were observed. However, at 1 198 and 1 223 K, secondary-recrystallized matrix grains could not be observed and at 1 373 K very few isolated grains could be observed. Therefore, data from 1 248 to 1 348 K are mainly used for discussion. The orientation relation between the central point of each isolated grain and the close position (around 10 μm) from the grain boundary in a matrix was measured.

Here, the two measurements of {100} pole figure and EBSD are mutually independent.

### Table 1. Chemical composition of steel used.

<table>
<thead>
<tr>
<th>Element</th>
<th>Mass%</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>0.001</td>
</tr>
<tr>
<td>Si</td>
<td>3.23</td>
</tr>
<tr>
<td>Mn</td>
<td>0.078</td>
</tr>
<tr>
<td>S</td>
<td>0.026</td>
</tr>
<tr>
<td>Al</td>
<td>0.029</td>
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<tr>
<td>N</td>
<td>0.0077</td>
</tr>
<tr>
<td>Sn</td>
<td>0.07</td>
</tr>
<tr>
<td>Cu</td>
<td>0.07</td>
</tr>
</tbody>
</table>

Fig. 1. Cross sectional microstructures for the transverse direction.

(a) Cross sectional microstructure extracted at 1248K

(b) Cross sectional microstructure extracted at 1273K

(c) Examples of colony grains

\[
V = \frac{dR}{dt} = AMP = AME(1/Rc - K/R ± KZ/A) \quad \ldots \ldots (1)
\]

$V$ = velocity of grain growth,
$R$ = radius of secondary grain,
$A$ = geometrical factor,
$M$ = mobility of a grain boundary,
$P$ = total driving force.
average grain boundary energy of primary matrix, 
Rc = critical radius of primary matrix grains, 
K = ratio of secondary grain boundary energy to primary grain boundary energy, 
Z = Zener term.

Here, regarding the sign of the term Z, if the noted grain is growing, the sign is minus, and if it is shrinking, the sign is plus.

The total driving force (P) consists of three components: the driving force derived from the grain boundary energy of primary matrix grains (P1), the retarding force derived from the shrinking force of the secondary grain itself (P2), and the retarding force derived from the pinning effect of second phase particles (P3). Small K (grain boundary energy) makes P2 and P3 small. As a result, V becomes large, i.e., lower grain boundary energy might cause faster grain growth in the case of the same mobility (M). On the contrary, for the faster V, M should be high and/or K should be low.

As isolated grains in the matrix are shrinking, Rc can be infinite, and Eq. (1) is rewritten as follows,

\[ V = \frac{dR}{dt} = AMP = AME(-K/R + KZ/A) = AME(-1/R + Z/A) \] ............................(2)
Figure 3 shows the \{100\} pole figure by EBSD for each temperature. The orientation of the matrix grains was not exact Goss orientation, to be exact \{540\}\(\langle\langle001\rangle\), because the preparation and setting conditions of the specimens might not be perfect. Although each cross section was carefully revealed, error is unavoidable. The orientation of each grain against the rolling direction and the normal plane is not always accurate, but the relation between two grains is sufficiently accurate and believable.

Figure 4 shows the frequency of CSL grain boundary \(\Sigma_i (i=1-29)\) regarding the misorientation between an isolated grain and a matrix grain for each extraction temperature. Here, 29 of the \(\Sigma\)-value means that it is more than 27 and it does not have a CSL relation by the Brandon criterion, hereinafter referred to as the general grain boundary (GGB).

In Fig. 2, it was already shown that \(\Sigma 3\) orientation to Goss in the primary texture is not as strong as \(\Sigma 9\) or \(\Sigma 5\). The CSL model is based on the assumption that \(\Sigma 9\) and \(\Sigma 5\) are the most mobile and more mobile grain boundaries, respectively. This result does not contradict this assumption.

In Fig. 4, the orientation of \(\Sigma 3\) has survived at lower temperature and at higher temperature GGB survived most frequently, and no isolated grains of \(\Sigma 9\) and \(\Sigma 5\) to Goss could be observed. This means the \(\Sigma 3\) grain boundary and GGB are not as mobile as other grain boundaries, especially \(\Sigma 9\) and \(\Sigma 5\). i.e., \(M\) (mobility) of \(\Sigma 9\) and \(\Sigma 5\) might be higher than that of \(\Sigma 3\) and GGB, and/or \(K\) (grain boundary energy) of \(\Sigma 9\) and \(\Sigma 5\) might be lower than that of them.

Incidentally, Wolf\(^{25}\) reported that the grain boundary energy of \(\Sigma 3\) might be lower than that of \(\Sigma 9\) and \(\Sigma 5\). If the grain boundary energy is more dominant than the mobility to the grain growth, the \(\Sigma 3\) boundary is rarely observed. Therefore, in this case, \(M\) might be more dominant than \(K\) and should have a large difference.

On the other hand, Nakayama et al.\(^{22}\) reported that instead of the difference of the mobility, with only that of the grain boundary energy, the secondary recrystallization can be simulated. Therefore, other possible causes, e.g., an interaction between inhibitors and mobility will be considered.

### 3.3. High Angle Misorientation

Figure 5 shows the misorientations between isolated grains and a matrix grain. Those from 50 to 60° show the highest frequency. By the Brandon criterion,\(^{26}\) a large part of them do not have the \(\Sigma 3\) relation. There are possibly two causes of this. One is that the isolated grain is not perfectly three-dimensionally isolated and the pure relation between an isolated grain and matrix could not be estimated. The other is that GGB has a special physical meaning from the point of view of grain boundary energy and mobility. But as the former can no longer be considered, only the latter is discussed.

Hutchinson et al.\(^{26}\) have already reported the same result and the boundaries with very high misorientation did not contribute to the recrystallization process due to low mobility. In their case, the material was pure iron, and the tensile stress and annealing process were applied in order to obtain island (isolated) grains. Furthermore, although the pinning effect (P3) was not dominant in the pure iron, that the same results were obtained suggests that the contribution of P3 might be small. As a result, \(M\) of \(\Sigma 3\) and GGB was lower than \(\Sigma 9\) and \(\Sigma 5\) and supports the above-mentioned results.

Furthermore, Fig. 6 shows the correlation between the frequency of \(\Sigma 3\) and GGB. At lower temperature, the frequency of \(\Sigma 3\) is higher than other relations. As the extraction temperature increased, the frequency of \(\Sigma 3\) decreased and that of GGB increased. The \(\Sigma 3\) boundary seems to change to GGB. That is, the most immobile grain boundary is GGB and the second most immobile grain boundary is \(\Sigma 3\) in the case of GO, whose lattice structure is BCC.

Incidentally, Wolf\(^{25}\) reported the grain boundary cusp at around 50° misorientation and its grain boundary energy is considerably low in \(\alpha\)-Fe. As the extraction temperature increased, the frequency of \(\Sigma 3\) decreased and that of this GGB increased. It seems as if the orientation corresponding to
to those cusps would survive. It is very interesting that the misorientation angle between the calculation and this observation is almost the same, although the relation is not defined as $\Sigma 11$. Therefore, new ideas will be necessary. As mentioned above, $M$ (mobility) of this GGB might be low. However, this lower grain boundary energy could also affect grain boundary migration, i.e., although the effects of $M$ and $K$ in Eq. (1) are mutually independent, an interaction between $M$ and $K$ considering inhibitors should be considered in order to explain this phenomena.28)

In addition, $\Sigma 1$ is not as mobile.27) Figure 4 also shows a few $\Sigma 1$ grain boundaries even at higher temperatures. This supports the well established character of $\Sigma 1$.

4. Conclusion

During secondary recrystallization of GO, isolated grains are often observed in secondary-recrystallized Goss grains (matrix). In an extraction experiment, the crystallographic relations between isolated grains and matrix grains were investigated.

(1) Although the $\Sigma 9$ grain boundary is the most frequent in the primary texture, it could not be observed in the relation, but specific grains, which have $\Sigma 3$ and a high angle misorientation relation, were often observed. As the extraction temperature increased, the frequency of $\Sigma 3$ decreased and that of GGB increased.

(2) It is concluded that the $\Sigma 9$ boundary is mobile, $\Sigma 3$ is less mobile and GGB is the least mobile. This does not contradict the assumption of the CSL model and supports it.

(3) Although the reason might be due to “mobility,” the contribution of grain boundary energy cannot be neglected owing to other studies. New ideas will be necessary.

REFERENCES