Mechanism of Orientation Selectivity of Secondary Recrystallization in Fe–3%Si Alloy

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Selective growth behavior of $\{110\}\langle 001 \rangle$ grains has been studied utilizing the temperature gradient annealing method. As grains grow, the average deviation angle from the ideal $\{110\}\langle 001 \rangle$ orientation becomes smaller and orientation distribution changes corresponding to that of coincidence grains in the matrix. Secondary recrystallization temperature depends on the orientation of secondary recrystallized grain and sharper $\{110\}\langle 001 \rangle$ grains grow preferentially at lower temperatures.

These phenomena are explained by modified Hillert's model of grain growth, which takes the grain boundary characteristics into account. Sharper $\{110\}\langle 001 \rangle$ grains, which have higher frequency of coincidence grains in the primary recrystallized matrix, suffer lower pinning force from the precipitates and thus grow preferentially at lower temperatures.

KEY WORDS: silicon steel; secondary recrystallization; texture; inhibitor; coincidence boundary; grain growth.

1. Introduction

Grain oriented silicon steel is a soft magnetic material widely used for transformer cores. Magnetic properties of the product are closely related to the sharpness of $\{110\}\langle 001 \rangle$ texture, *i.e.* Goss texture, which is evolved by secondary recrystallization. Iron crystal has an anisotropy in magnetization and the axis of easy magnetization is $\langle 001 \rangle$. If the sharp $\{110\}\langle 001 \rangle$ texture is developed, the product has a superior magnetic properties, such as low core loss, low magnetostriction and high permeability along the rolling direction.

The principal process for developing Goss texture was invented by Goss in 1934^{1,2)} and was industrialized by ARMCO in the 1940's [CGO: conventional grain oriented silicon steel].³⁾ The average deviation angle of $\langle 001 \rangle$ axis is 7 degrees from the rolling direction. High permeability grain oriented silicon steels (average misorientation of 3 to 4 degrees) were developed mainly by NSC in the 1960's [HI-B].^{4,5)} The major differences between these processes are the following two features:

(1) primary recrystallization texture; the CGO process involves two stage cold rolling with an intermediate recrystallization annealing and the HI-B process a single stage cold rolling with heavy reduction of about 87% compared to the final reduction of about 50% in the CGO process. Depending on the difference of these final cold rolling reduction, the $\{110\}\langle 001 \rangle$ and $\{111\}\langle uvw \rangle$ are major components of the primary recrystallization texture in the CGO process. On the other hand, $\{110\}\langle 001 \rangle$ is a minor component and $\{111\}\langle 112 \rangle$

is a major component in the HI-B process.^{6–8)}

(2) inhibitor; CGO utilizes MnS and HI-B AlN as a main inhibitor in addition to MnS for the grain growth inhibitor of primary recrystallized matrix.

Many studies have been reported on secondary recrystallization in Fe–3%Si alloy. Some researchers^{9–11)} investigated the origin of Goss nuclei and claimed that the size advantage of the nuclei takes an important role in the evolution of Goss texture. Most of these researches were related to the CGO type process. On the other hand, in the case of HI-B type process, others^{12–16)} investigated the relationship between the secondary recrystallization texture and primary recrystallization texture and claimed that some special boundaries (for example, $\Sigma 9$ coincidence boundaries) take an important role in the evolution of Goss texture.

Present authors have studied the effect of cold rolling reduction on primary and secondary recrystallization texture evolution and have found that the frequency of coincidence boundary increases with cold rolling reduction.¹⁷⁾ This fact suggests that the oriented nucleation mechanism dominates the preferred growth of Goss grains in the CGO type process and the oriented growth mechanism in the HI-B type process, which is industrially more effective for the texture control. In this consequence, the present authors have also found that the sharpest Goss texture (average misorientation of 1 degree or so) evolves when grain growth of secondary recrystallization is accelerated by temperature gradient annealing in the HI-B process.¹⁸⁾ However, the precise observation of grain growth behavior in the temperature gradient an-

nealing has not been performed.

The purpose of this paper is to investigate the grain growth behavior of secondary recrystallized Goss grains in the temperature gradient annealing and clarify the mechanism of orientation selectivity in the secondary recrystallization process.

2. Experimental Procedure

The starting material was Fe-3%Si primary recrystallized sheet of 0.2 mm thickness with the following chemical compositions; Si: 3.3, Mn: 0.1, C: 0.002, Al: 0.03, N: 0.007 mass%. The processing steps involved a hot rolling, an annealing, a single stage cold rolling with the reduction of 90% and a primary recrystallization and decarburization annealing. In order to inhibit the grain growth of primary recrystallized matrix effectively, nitriding treatment was performed to increase the nitrogen content to 0.05 mass%.¹⁹⁾ The sheet was then heated up to 1200°C with the heating rate of 10°C/h in the temperature gradient of 0.5°C/mm perpendicular to the rolling direction in 90%N₂-10%H₂ mixture gas.

Orientations of the secondary recrystallized grains were measured by back-reflection Laue diffraction method. The complete {100} pole figures were measured by X-ray diffraction measurement from the surface (1/5th of the thickness) and center layer of the specimen. Primary recrystallization texture was determined by three dimensional texture analysis by vector method²⁰⁾ and then orientation relationships between the secondary recrystallized grains and primary recrystallization texture were analyzed.

3. Results and Discussions

3.1. Texture Evolution during Grain Growth

Figure 1 is an optical macrograph which shows the macrostructure of the specimen after the secondary recrystallization annealing and Fig. 2 illustrates the macrostructure of the secondary recrystallized grains. It can be seen from these figures that several secondary recrystallized grains nucleated at the edge of the specimen and grew along the temperature gradient direction. It has been confirmed that the grains grow along the temperature gradient direction regardless of the cold rolling direction. As grains grew, some of the grains grew preferentially to form a columnar structure. The secondary recrystallized grains are classified into two groups; stunted grains and growth grains. Orientation of individual secondary recrystallized grain was measured and represented as the deviation angles from the ideal Goss orientation around the three axes; normal direction of the sheet (ND), rolling direction (RD), and transverse direction (TD). It can be seen from Fig. 3 that the orientation distributions of these two groups were different. The deviation angles of the growth grains were smaller than those of the stunted grains and the orientation distribution of the growth grains was qualitatively estimated as the RD-deviated type ($\Delta RD > \Delta ND \sim \Delta TD$), on the other hand that of the stunted grains was the ND-deviated type ($\Delta ND > \Delta RD > \Delta TD$). These facts

Fig. 1. Macrostructure of secondary recrystallized grains in the temperature gradient annealing.

Fig. 2. Illustration of macrostructure of secondary recrystallized grains in the temperature gradient annealing.

suggest that two modes of preferential growth of Goss grains operate in the process and selective growth during grain growth stage takes a more important role in controlling the Goss texture evolution than that in the nucleation stage.

In order to find out the role of primary recrystallization texture, primary recrystallization texture was measured and analyzed to evaluate the orientation distribution of Goss nuclei and that of coincidence ($\Sigma 9$) grains in the matrix. Figures 4(a) and 4(b) show the complete {100} pole figures of the primary recrystallized specimen. Figure 5 shows the three dimensional orientation distribution of the center layer calculated from Fig. 4(b). It can be seen that in the primary recrystallization texture the Goss component is a minor component and major components are classified as follows; (a) γ -fiber with the peak of $\{111\}\langle 112 \rangle$ and (b) rotated α -fiber, which is rotated about 20 degrees around the ND axis, with the peak of $\{411\}\langle 148 \rangle$. These two peaks, $\{111\}\langle 112 \rangle$ and $\{411\}\langle 148 \rangle$, are calculated to have a $\Sigma 9$ coincidence relationship to Goss orientation assuming the Brandon criterion²¹⁾ and therefore they are important for the Goss

Fig. 3. Change of orientation distribution of secondary recrystallized grains during grain growth.

Fig. 4. Complete $\{100\}$ pole figures of primary recrystallization texture; (a) surface layer, (b) center layer.

grain growth. It has been reported that Goss grains nucleate in the surface layer,^{9,10)} hence, estimation of the orientation distribution of Goss nuclei was carried out by the pole figure of the surface layer and that of coincidence grains by the average pole figure of the both

Fig. 5. Three dimensional orientation distribution of primary recrystallization texture.

Fig. 6. Orientation distribution of $\{110\}\langle 001 \rangle$ nuclei and that of $\Sigma 9$ coincidence grains in the primary recrystallization texture.

layers. It can be seen from Fig. 6 that the orientation distribution of Goss nuclei and that of coincidence grains are quite different. Orientation distribution of Goss nuclei have a concave shape and that of coincidence grains a convex shape and they are qualitatively estimated as the ND-deviated type ($\Delta ND > \Delta RD > \Delta TD$) and the RD-deviated type ($\Delta RD > \Delta TD \sim \Delta ND$), respectively.

Thus, with the comparison between the orientation distribution of secondary recrystallized grains and those of Goss nuclei and coincidence grains in the primary recrystallized matrix, it is concluded that the orientation distribution of secondary recrystallized grains corresponds to that of Goss nuclei at the initial stage of secondary recrystallization and it changes successively according to that of coincidence grains in the matrix as

Fig. 7. Optical micrograph of interface between the secondary recrystallized grains and primary recrystallized matrix.

grains grow.

3.2. Orientation Selectivity Behavior during Grain Growth

As shown in Fig. 1, sharper Goss grains which have higher frequencies of coincidence grains in the matrix were selected at the initial stage of grain growth among the several grains which were nucleated at the sheet edge, and after that they grew stationary to form a columnar structure. For the investigation of this selective growth mechanism, some specimens were quenched in the heating stage of the annealing. **Figure 7** is an optical micrograph which shows the interface structure between the secondary recrystallized grain and primary recrystallized matrix. The growth front of the secondary recrystallized grain is macroscopically uniform though it has some fluctuation in microscopic point of view. **Figures 8(a)** and **8(b)** illustrate the macrostructures of the growth front of the secondary recrystallized grains. It can be seen from these figures that secondary recrystallization temperature, which is defined as the growth front temperature, takes a specific value depending on the orientation of the secondary recrystallized grain. The secondary recrystallization temperatures of the secondary recrystallized grains in these specimens were in the range from 1050 to 1065°C. Secondary recrystallization temperatures of the sharper Goss grains were around 1050°C, which were about 10°C lower than those of the deviated Goss grains. Depending on the difference of secondary recrystallization temperature, sharper Goss grains grew preferentially by expanding the growth front and consuming the primary matrix region earlier than the deviated Goss grains. In the end, sharper Goss grains, which had the same secondary recrystallization temperature, grew stationary along the temperature gradient direction.

It is concluded that the secondary recrystallization temperature is an important parameter which dominates the selective growth behavior.

Fig. 8. Macrostructures of the growth front of the secondary recrystallized grains and orientation distribution of secondary recrystallized grains.

Fig. 9. Schematic illustration of the forces which affect the secondary recrystallized grain.

3.3. Mechanism of Orientation Selectivity

Secondary recrystallization phenomena are described by the modified Hillert's model,^{14,22,23)} which takes the textural effect into account. Grain growth rate of the secondary recrystallized grain is described by the following equation.

$$dR/dt = MP \dots\dots\dots (1)$$

$$P = AE\{(1/Rc) - (K/R) - (KZ/A)\} \dots\dots\dots (2)$$

where R : radius of secondary recrystallized grain, Rc : critical radius of primary recrystallized matrix grains, M : mobility, P : total driving force of secondary recrystallized grain, A : geometrical factor, E : average grain boundary energy of primary recrystallized matrix, K : ratio of secondary recrystallized grain boundary energy to that of matrix primary recrystallized grain boundary energy (E), Z : Zener term.

As can be seen from Eq. (2) and the diagram (Fig. 9), total driving force of secondary recrystallized grain consists of three components. The first (P1) is the driving force derived from the tension of grain boundaries of the primary recrystallized matrix. The second (P2) is the retarding force derived from the shrinking force of the secondary recrystallized grain itself. The third (P3) is the retarding force by the pinning effect of second phase particles.

It can be deduced from Eq. (2) that driving force from the grain boundary tension of the matrix grains takes the same value irrespective of the orientation of the secondary recrystallization grains. Two modes of preferred grain growth are recognized as; (1) "oriented nucleation derived from the size effect" (second term) and "oriented growth derived from the inhibition effect" (third term).

In the "nucleation stage" of secondary recrystallization, where grain size of secondary recrystallized grain takes the same order of magnitude compared with that of primary recrystallized matrix grains, the second component of the Eq. (2) takes an important role. Then, size and number of Goss nuclei dominate the preferred grain growth behavior at the initial stage of grain growth because this "size advantage effect" is reversely proportional to the grain size.

On the other hand, in the "grain growth stage", where grain size of the secondary recrystallized grain becomes

Fig. 10. Transmission electron micrograph which shows the pinning effect of precipitates.

Fig. 11. Change of nitrogen content during in the seating stage of annealing.

quite large compared with that of primary recrystallized matrix grains, the second term becomes negligible and the third component of Eq. (2) which represents "inhibition effect" dominates the grain growth behavior. Then, the primary recrystallized grains which have coincidence relationship to secondary recrystallized grain takes an important role. It has been reported that the coincidence boundary (or special boundary) has a lower grain boundary energy than the general boundary. Hence, the sharper Goss grain which has a higher frequency of coincidence grains to primary recrystallized matrix takes the lower statistical grain boundary energy (KE). Pinning force derived from the precipitates is estimated by the interaction energy between the grain boundary and precipitates, which is evaluated by the product of statistical grain boundary energy of the secondary recrystallized grain and area of cross section decreased by the second phase particles (Zener term). Thus, it is deduced that sharper Goss grains suffer lower pinning force from precipitates.

In the manufacturing process of grain oriented silicon steel, one of the requisites for secondary recrystallization is the presence of fine precipitates to inhibit the grain growth of matrix grains.²⁴⁾ As is demonstrated in Fig. 10, nitrides such as $(Al, Si)N$ and $(Si, Mn)N$ stagnate the grain boundary migration and affect as inhibitors in this system. Figure 11 shows the change of nitrogen content of the specimen measured by chemical analysis. As temperature rises, these nitrides dissolve and nitrogen

Fig. 12. Schematic diagram of orientation selectivity during grain growth of secondary recrystallization.

removes from the silicon iron sheet as nitrogen gas. Therefore, the nitrogen content which indicates inhibition effect decreases with increasing temperature. It can be seen that secondary recrystallization proceeds during the deteriorating stage of inhibition effect, because secondary recrystallization temperatures are in the temperature range from 1050 to 1065°C. Secondary recrystallization temperature is the onset temperature that secondary grain starts to grow by breaking away the pinning force by the second phase particles. As is shown in Fig. 8, secondary recrystallization temperature of sharper Goss grains are lower than those of deviated Goss grains. This fact suggests that the sharper Goss grains start to grow at the stage of higher inhibition level due to the grain boundary characteristics.

As a summary, the mechanism of selective growth of sharp Goss grain in the secondary recrystallization process is schematically illustrated in Fig. 12. Precise Goss grain, which has higher frequency of coincidence grains in the matrix, suffers lower pinning effect and starts to grow at lower temperatures and thus grows preferentially by expanding its growth front into the deviated Goss grains.

The present authors have proposed the same model of secondary recrystallization from the experimental results of *in situ* observation of secondary recrystallization by synchrotron X-ray topography.^{25,26} *In situ* observation showed that $\{110\}\langle 001 \rangle$ grain which has higher frequency of coincidence grains in primary recrystallized matrix starts to migrate at lower temperature compared with the $\{110\}\langle 115 \rangle$ grain by breaking away the inhibition effect from second phase particles.

When the temperature gradient annealing was applied to the CGO type process, the selective growth behavior which is described in this paper was not observed. This result suggests that sharp orientation distribution of coincidence grains in the primary recrystallized matrix, which is the characteristic feature of the primary recrystallization texture in the HI-B type process, is indispensable for the selective growth of Goss grains.

4. Conclusions

Orientation selectivity of Goss grains in the secondary recrystallization process were investigated in the tem-

perature gradient annealing.

(1) Sharp Goss grains selectively grow during the grain growth process; the average deviation angle from the ideal Goss orientation becomes smaller and orientation distribution changes corresponding to that of coincidence grains in the primary recrystallized matrix.

(2) Secondary recrystallization temperature is a dominant parameter for selective growth. Sharper Goss grains grow at lower temperatures and expand their territories in the primary recrystallized matrix.

(3) Selective growth behavior is explained by the modified Hillert's model of grain growth which takes the grain boundary characteristics into account. Sharper Goss grains, which have higher frequency of coincidence grains in the primary matrix, suffer lower pinning effect from precipitates and thus grow preferentially at lower temperatures.

(4) Selective growth of sharp Goss grains takes a dominant role of texture control in the HI-B process, which is characterized by the sharp orientation distribution of coincidence grains in the primary recrystallization texture.

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