Magnetic Properties and Recrystallization Texture Evolutions of Phosphorus-bearing Non-oriented Electrical Steel Sheets

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The effect of phosphorus (P) on magnetic properties and also on recrystallization texture of non-oriented electrical steel sheets has been investigated to develop core materials with low core loss and high permeability. Specimens containing different amounts of P were cold-rolled to various thicknesses, i.e., with various cold-rolling reductions, and annealed for recrystallization and grain growth. Magnetic induction of the steel with a large amount of P was higher than that of the steel with a small amount of P. Moreover, magnetic induction of the steel with a large amount of P slightly decreased with reduction of sheet thickness, i.e., with an increase in cold-rolling reduction, whereas magnetic induction of the steel with a small amount of P dramatically decreased. The most effective way to reduce core loss was to reduce thickness of electrical steel sheets. Therefore, P-bearing thin gauge non-oriented electrical steel sheets achieved low core loss and high permeability. The typical magnetic properties of P-bearing non-oriented electrical steel sheets with a thickness of 0.27 mm were 16.6 W/kg in W10/400 and 1.73 T in B50. These excellent magnetic properties were provided by the recrystallization texture control by P. {111}(225) component in recrystallization texture, which deteriorates magnetic properties of electrical steel sheets, was suppressed during recrystallization. Furthermore, {25°, 10–15°, 45°} component significantly developed at the expense of {111}(112) component during grain growth. P segregation at initial grain boundaries would be responsible for this texture evolution. Accordingly, P would greatly contribute to the improvement of magnetic properties of non-oriented electrical steel sheets through the recrystallization texture control.

KEY WORDS: core loss; magnetic induction; recrystallization; texture; phosphorus; non-oriented electrical steel sheet.

1. Introduction

Non-oriented electrical steel sheets have been widely used for core materials of motors. Most important properties of these sheets are core loss and permeability. Recently, there has been a great demand for reducing core loss, especially high-frequency core loss, of non-oriented electrical steel sheets from the viewpoint of energy conservation and prevention of global warming. The most effective solution to reduce high-frequency core loss is to reduce thickness of electrical steel sheets, so that thin gauge non-oriented electrical steel sheets of 0.35 mm or less in thickness have been used for high-efficiency motors.1,2 However, low core loss is incompatible with high permeability in conventional non-oriented electrical steel sheets, since magnetic induction of these sheets is dramatically diminished with an increase in cold-rolling reduction due to a recrystallization texture change.3,4 Therefore, texture control is significantly important for non-oriented electrical steel sheets, and accordingly, effects of the typical elements which tend to segregate at grain boundaries such as Sb,4–7 Sn8–10 and P11,12 on recrystallization texture of non-oriented electrical steel sheets have been investigated. Shimanaka et al.4 reported that near {100}/(0vw) component is developed by Sb addition. Lyukdkovsky et al.5 reported that Sb promotes the growth of {110} and {100} component at the expense of {111} and {211} component. Vodopivec et al.6 reported that {111} component is greatly diminished by Sb addition. Takashima et al.7 reported that {111} component is suppressed by Sb addition. Kubota et al.8 reported that {111} component decreases and {110} component increases with Sn addition in both non skin-pass and skin-pass processes. Furthermore, Kubota et al.9 reported that {411} and {100} component increases with an increase in the amount of Sn through cold-rolling reduction range higher than 80%. Goddec et al.10 reported that {100}/(001) component increases, whereas {111} component decreases with Sn addition. On the contrary to these beneficial effects of Sb4–7 and Sn8–10 on recrystallization texture, it has been reported that {111}(112) component is developed, and therefore magnetic induction decreases with an increase in the amount of P.11,12 It is well-known that P improves drawability of low carbon steels, therefore effects of P on recrystallization texture of low carbon steels have been investigated.13,14 Inagaki et al.15 reported that {554}/(225) component increases with an increase in the amount of P. These results are ob-
tained through the experiment without a hot-rolled band annealing, and consequently grain diameters before cold-rolling decreases with an increase in the amount of P. Hence, it would seem that the development of \{111\}(112) and \{554\}(225) components by P addition results from the decrease in grain diameters before cold-rolling. Therefore, the effect of P on magnetic properties and also on recrystallization texture has not been clarified yet. In this study, the effect of P on magnetic properties of non-oriented electrical steel sheets has been investigated to develop core materials with low core loss and high permeability. Furthermore, texture evolution of P-bearing non-oriented electrical steel sheets during recrystallization and grain growth has been investigated. As a result, magnetic induction of the steel with a large amount of P was higher than that of the steel with a small amount of P, and moreover, magnetic induction of P-bearing non-oriented electrical steel sheet slightly decreases with an increase in cold-rolling reductions due to a recrystallization texture change.

2. Experimental

The chemical composition of steels used in the present study is given in Table 1. The amount of P in 0.01P steel would correspond to that of conventional high-grade non-oriented electrical steel sheets. Hot-rolled bands of these steels with a thickness of 2.0 mm were heated to 680, 800 and 900°C at a rate of 40°C/h, kept for 10 h in an Ar atmosphere and cooled to the room temperature at a rate of 40°C/h. These specimens were cold-rolled at a reduction of 75, 83 and 87% to 0.5, 0.35 and 0.27 mm in thickness, respectively. Then they were annealed at 1000°C for 30 s in an Ar atmosphere for recrystallization and grain growth, hereinafter called as a final-anneal. Transmission electron microscopy (TEM) observation with blank-replica method was carried out after the hot-rolled band annealing. Magnetic measurements were carried out for final-annealed sheets by a 30 mm in width and 100 mm in length single strip tester both in rolling and transverse directions. The measured values were averaged to parallelize with Epstein strip tester both in rolling and transverse directions. The sheets by a 30 mm in width and 100 mm in length magnetic measurements were carried out for final-annealed steels.

For the investigation into the effect of P segregation at initial grain boundaries on texture evolution during recrystallization and grain growth in the final-annealing, hot-rolled bands of 0.1P steel listed in Table 1 with a thickness of 2.0 mm were used. Here, initial means before cold-rolling. Hot-rolled bands of 0.1P steel were heated to 825°C at a rate of 40°C/h, kept for 10 h in an Ar atmosphere and cooled to the room temperature at a rate of 40°C/h. Specimens were again annealed at 900°C for 2 min in a salt bath, and air-cooled to the room temperature in order to eliminate the grain boundary segregation of P. Additionally, some specimens were annealed at 700°C for 100 h in an Ar atmosphere to sufficiently promote the grain boundary segregation of P and then quenched into water. Hereinafter, this heat treatment is called as a segregation treatment. Specimens with and without the segregation treatment were cold-rolled at a reduction of 87% to 0.27 mm in thickness. Then they are heated at a rate of 20°C/s and cooled at a rate of 20°C/s immediately after being reached the temperature of interest or annealed at 1000°C for 30 s in an Ar atmosphere. Optical microscope observation was carried out for specimens before and after the segregation treatment. Textures of cold-rolled and annealed sheets were determined in the same way described above.

3. Results

3.1. Magnetic Properties

The effect of cold-rolling reduction and thickness on magnetic properties is shown in Fig. 1. Core loss of both steels decreased with reduction of sheet thickness and magnetic induction also decreased with an increase in cold-rolling reduction. However, magnetic induction of 0.1P steel was higher than that of 0.01P steel at any cold-rolling reduction. Furthermore, magnetic induction of 0.1P steel slightly decreased with an increase in cold-rolling reduction, whereas magnetic induction of 0.01P steel was dramatically reduced. These results mean that P-bearing thin gauge non-oriented electrical steel sheets achieved low core loss and high permeability. The typical magnetic properties of 0.1P steel with a thickness of 0.27 mm were 16.6 W/kg in W10/400 and 1.73 T in B50. It is reported that electrical resistivity of α-Fe significantly increases with P content. However, the effect with 0.1 mass% P is approximately $1 \times 10^{-3}$ Ω·m. Hence, the beneficial effect of P on core loss presented in Fig. 1 should be attributed to the decrease in hysteresis loss due to the texture improvement mentioned below as well as the decrease in eddy current loss.

| Table 1. Chemical composition of steels. (mass%). |
|----------|-------|-------|-------|-------|-------|
| Steels   | C     | Si    | Mn    | P     | sol.Al |
| 0.01P    | 0.0019| 1.99  | 0.24  | 0.013 | 0.29  |
| 0.1P     | 0.0019| 2.07  | 0.21  | 0.099 | 0.29  |

Fig. 1. Effect of cold-rolling reduction and thickness on magnetic properties. Hot-rolled band annealing temperature is 800°C.
3.2. Recrystallization Texture

The effect of (111)/ND pole intensity on magnetic induction is shown in Fig. 2. Magnetic induction decreased with an increase in (111)/ND pole intensity. Moreover, (111)/ND pole intensity of 0.1P steel would be lower than that of 0.01P steel. Recrystallization textures of 0.27 mm thick steel after the final-annealing are presented in Fig. 3. {111}(112) intensity of 0.1P steel was lower than that of 0.01P steel. Furthermore, the intensity of \( \{ \phi_1, \phi_2 \} = \{ 25^\circ, 5^\circ-15^\circ, 45^\circ \} \), which is comparatively close to \{100\} plane, of 0.1P steel was higher than that of 0.01P steel. Therefore, this texture change was likely to have caused the increase in magnetic induction with P content shown in Fig. 1. {111}(112) components seem to be suppressed by P.

Optical microstructures of 0.1P steel before and after the segregation treatment are shown in Fig. 4. It can be seen that grain diameters remained unchanged during the segregation treatment. Consequently, specimens with and without the segregation treatment had the same amount of P and also the same initial grain diameter, but the different state of P segregation at initial grain boundaries mentioned below.

Reconstitution textures of 0.1P steel with and without the segregation treatment after being annealed at 1000°C for 30 s are presented in Fig. 5. {111}(112) intensity of 0.1P steel with the segregation treatment was much lower than that of 0.1P steel without the segregation treatment. Furthermore, the intensity of \( \{ \phi_1, \phi_2 \} = \{ 25^\circ, 10^\circ-15^\circ, 45^\circ \} \) of 0.1P steel with the segregation treatment was higher than that of 0.1P steel without the segregation treatment. Recrystallization texture of 0.1P steel without the segregation treatment was found to be fairly close to that of 0.01P steel shown in Fig. 3. This indicates that recrystallization texture is hardly influenced only by P addition.

3.3. Texture Evolution during Recrystallization

Texture evolutions of 0.1P steel with and without the segregation treatment during recrystallization are presented in Fig. 6. As can be seen in the figures, both steels showed almost the same cold-rolled texture; main component was \( \alpha \)-fiber from \{100\}(011) to \{111\}(011). Moreover, textures of both steels remained unchanged until 700°C. Subsequently, they have dramatically changed at 750°C. {111}(112) intensity suddenly increased, whereas \{111\}(011) intensity significantly decreased. It should be pointed out that, {111}(112) intensity of 0.1P steel with the segregation treatment was lower than that of 0.1P steel without the segregation treatment at 750 and at 800°C, and the intensity of \( \alpha \)-fiber from \{100\}(011) to \{211\}(011) of 0.1P steel with the segregation treatment was higher than that of 0.1P steel without the segregation treatment at 800°C.

3.4. Texture Evolution during Grain Growth

Texture evolutions of 0.1P steel with and without the segregation treatment during grain growth are presented in Fig. 7. The change in intensity of \{111\}(112) and \( \{ \phi_1, \phi_2 \} = \{ 25^\circ, 15^\circ, 45^\circ \} \) with temperature is shown in Fig. 8. As can be seen in these figures, textures of 0.1P steel without the segregation treatment remained almost unchanged during grain growth. On the other hand, in 0.1P steel with the segregation treatment, \{111\}(112) intensity dramatically dropped, and furthermore, \( \{ \phi_1, \phi_2 \} = \{ 25^\circ, 10^\circ-15^\circ, 45^\circ \} \) component significantly developed during grain growth at 950°C or more, especially at 1000°C for 30 s. Besides, \{111\}(112) intensity of 0.1P steel with the segregation...
treatment was lower than that of 0.1P steel without the segregation treatment at any temperature.

4. Discussion

4.1. Estimation of Phosphorus Concentration at Grain Boundary

As mentioned above, P is one of the typical elements which segregate at grain boundaries. According to TEM observation, no precipitate containing P was detected after the hot-rolled band annealing. This would imply that the texture change shown in Fig. 5 is due to P segregation at initial grain boundaries. To verify this idea, P concentration at initial grain boundaries in specimens with and without the segregation treatment should be estimated. Time evolution of grain boundary coverage at given temperature can be described as, 

\[ \text{Intensity level is the same as Fig. 5.} \]

\[ \text{Fig. 6. } \phi_2=45^\circ \text{ ODF of 0.1P steel (A), (B), (C), (D) without and (A'), (B'), (C'), (D') with the segregation treatment at the mid-plane of 0.27 mm thick sheets. (A)(A') as cold-rolled, (B)(B') 700°C, (C)(C') 750°C, (D)(D') 800°C. Intensity level is the same as Fig. 5.} \]

\[ \text{Fig. 7. } \phi_2=45^\circ \text{ ODF of 0.1P steel (A), (B), (C), (D) without and (A'), (B'), (C'), (D') with the segregation treatment at the mid-plane of 0.27 mm thick sheets. (A)(A') 850°C, (B)(B') 900°C, (C)(C') 950°C, (D)(D') 1000°C. Intensity level is the same as Fig. 5.} \]

\[ \text{Fig. 8. The change in intensity of } \{111\}(112) \text{ and } \{\phi_1, \phi_2\} = \{25^\circ, 15^\circ, 45^\circ\} \text{ with temperature. ("As CR" indicates "as cold-rolled").} \]
concentration during the each heat treatment is estimated. Time evolution of P segregation at grain boundaries in 0.1P steel at 825°C is shown in Fig. 9. Initial grain boundary coverage of P is unknown, thus \( X_0 \) is assumed to be zero in this estimation. As can be seen in the figure, grain boundary coverage of P reaches the equilibrium value at 825°C after 0.1 h even on the assumption that \( X_0 \) is zero; therefore, grain boundary coverage could sufficiently reach the equilibrium value at 825°C, which is equal to 0.30, within 10 h. In the present study, specimens annealed at 825°C were cooled at a rate of 40°C/h and consequently the grain boundary segregation could proceed further during cooling process. Therefore, \( X_0 \) would be at least 0.30 in the following annealing process by a salt bath.

Time evolution of P segregation at grain boundaries in 0.1P steel at 900°C is shown in Fig. 10. In consideration of the grain boundary segregation during cooling process from 825°C, \( X_0 \) is taken to be the equilibrium value at 400, 600, 700, as well as at 825°C, which is equal to 0.93, 0.63, 0.46 and 0.30, respectively. Grain boundary coverage of P reaches the equilibrium value at 900°C after 10 s even on the assumption that \( X_0 \) is 0.93, therefore, grain boundary coverage could reach the equilibrium value at 900°C, which is equal to 0.23, within 2 min. The grain boundary segregation during air-cooling process is negligible, and accordingly \( X_0 \) would be 0.23 in the following segregation treatment. This estimation indicates that P concentration at initial grain boundaries in the specimen without the segregation treatment is 0.23.

Time evolution of P segregation at grain boundaries in 0.1P steel at 700°C is shown in Fig. 11. Grain boundary coverage of P could reach the equilibrium value at 700°C, which is equal to 0.46, within 10 h. Consequently, P concentration at initial grain boundaries of specimens with the segregation treatment would be twice as large as that of specimens without the segregation treatment. This estimation would imply that texture evolution shown in Figs. 5–7 is due to the difference of P concentration at initial grain boundaries.

### 4.2. Texture Evolution of Phosphorus-bearing Non-oriented Electrical Steel Sheets

Figure 6 shows that \{111\}(112) intensity of 0.1P steel with the segregation treatment was lower than that of 0.1P steel without the segregation treatment, and in addition, intensity of \( \alpha \)-fiber from \{100\}(011) to \{211\}(011) in 0.1P steel with the segregation treatment remained high level during recrystallization. Generally, \{111\} recrystallized grains arise near initial grain boundaries.\(^{22,23}\) Therefore, it is indicated that in the case where P segregation at initial grain boundaries was sufficiently promoted, \{111\}(112) component was suppressed during recrystallization. Additionally, \( \alpha \)-fiber from \{100\}(011) to \{211\}(011) would remain until full-recrystallization due to the suppression of \{111\}(112) component.

Figures 5, 7 and 8 show that \( \{ \varphi_1, \Phi, \varphi_2 \} = \{25°, 10–15°, 45°\} \) component dramatically developed, whereas \{111\}(112) component significantly decreased in 0.1P steel with the segregation treatment during grain growth. To consider the texture evolution of 0.1P steel with the segregation treatment during grain growth, the origin of \( \{ \varphi_1, \Phi, \varphi_2 \} = \)
seen that the rotation axis is close to the rotation angle is 40.30°. This relationship is close to grain boundaries between {111} and {100} that {25°, 10–15°, 45°} should be examined. Homma et al.\(^2\) reported that \(\{h \!, 1, 1\!/h \!, 1, 2\!\) recrystallized grains arise from {100} to {211} recrystallization-retdarded \(\alpha\)-fiber texture. \(\{\varphi_1 \!, \Phi \!, \varphi_2\!\} = \{25°, 10–15°, 45°\}\) component is close to {411} which belongs to \(\{h \!, 1, 1\!/h \!, 1, 2\!\) fiber texture. Furthermore, \(\alpha\)-fiber from {100} to {211} remained until full-recrystallization in 0.1P steel with the segregation treatment. Therefore, it is probable that \(\{\varphi_1 \!, \Phi \!, \varphi_2\!\} = \{25°, 10–15°, 45°\}\) component would arise from {100} to {211} recrystallization-retdarded \(\alpha\)-fiber texture after the recrystallization of {111} component, as well as {411} component.

The orientation relationship between {111} and \(\{\varphi_1 \!, \Phi \!, \varphi_2\!\} = \{25°, 15°, 45°\}\) is calculated from Euler-space coordinates. The rotation axis is shown in Fig. 12. It can be seen that the rotation axis is close to {011}, and moreover, the rotation angle is 40.30°. This relationship is close to \(\Sigma\) coincidence site lattice (CSL) boundary. It is reported that CSL boundaries have higher migration mobility than other general boundaries.\(^2\) On the basis of this idea, grain boundaries between {111} and \(\{\varphi_1 \!, \Phi \!, \varphi_2\!\} = \{25°, 10–15°, 45°\}\) have a priority in grain boundary migration under the solute drag effect of P. Consequently, \(\{\varphi_1 \!, \Phi \!, \varphi_2\!\} = \{25°, 10–15°, 45°\}\) component could develop at the expense of {111} recrystallized grains during grain growth due to its advantageous orientation relationship in grain boundary migration.

Hence, P segregation at initial grain boundaries would be responsible for the beneficial recrystallization texture evolution in non-oriented electrical steel sheets. P has beneficial effects on recrystallization texture as well as Sb\(^{1-7}\) and Sn\(^{8,9}\) and thus it would greatly contribute to the improvement of magnetic properties of non-oriented electrical steel sheets.

5. Conclusion

Effect of P on magnetic properties and also on recrystallization texture of non-oriented electrical steel sheets has been investigated. The following results were obtained.

(1) Magnetic induction of the steel with a large amount of P was higher than that of the steel with a small amount of P. Moreover, magnetic induction of the steel with a large amount of P slightly decreases with an increase in cold-rolling reduction, whereas magnetic induction of the steel with a small amount of P dramatically decreased.

(2) In recrystallization textures, \{111\} intensity of the steel with a large amount of P is lower than that of the steel with a small amount of P. This texture change corresponds to the magnetic induction change with P content.

(3) Although the steel has a large amount of P, recrystallization texture without the segregation treatment is fairly close to that of the steel with a small amount of P. Therefore, P segregation at initial grain boundaries would be responsible for the beneficial recrystallization texture evolution in non-oriented electrical steel sheets.

Appendix.

For the reference, some components are illustrated on \(\phi_2 = 45°\) ODF in Fig. A1.

**REFERENCES**