Effect of Nitriding on Grain Oriented Silicon Steel Bearing Aluminum

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All high permeability GO is manufactured using AlN as the main inhibitor. Two types of inhibitor preparation of GO have been realized industrially, i.e., extra high and extra low slab reheating temperature methods. Therefore, from a purely metallurgical viewpoint, the possibility of a middle temperature slab reheating method was examined and the effect of nitriding was investigated.

Metallurgically, this method is located between the Extra high and Extra low temperature methods. Furthermore, it is essential that a suitable combination of inhibitor intensity and primary diameter, controlled by nitriding and heat treatments respectively can provide sharp Goss orientation.

KEY WORDS: electrical steel; silicon steel; solid solution; recrystallization.

1. Introduction

Grain Oriented Silicon Steel (GO) is 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, along the rolling direction are closely related to the secondary recrystallization texture, i.e., the sharpness of \{110\}(001) (Goss texture). Therefore, it is essential to enhance the sharpness of the Goss texture.

The magnetic anisotropy of an Fe-single crystal was discovered in 1926.\(^2\) In 1934, the principal production process of GO was invented by Goss.\(^3\) Since then, much effort has been made to improve this material.\(^4,5\) As a result, the average deviation angle of the (001) axis from the rolling direction has been improved from 7 degrees to 3 degrees in Japan,\(^6,11\) referred to as high permeability grade. Furthermore, the slab reheating temperature can be decreased from an extra high temperature to a normal one.\(^12\) Both quality and production technology have been improved.

The production technology for the preparation of “inhibitors” can be classified into two categories.\(^12\) One is the ordinary technology where slabs are reheated over the solution temperatures of the inhibitor substances (such as MnS, AlN and MnSe) in order to cause them to disperse finely regardless of inhibitor type. The other is the new nitriding technology, developed by Nippon Steel,\(^12,13\) where slabs are reheated at sufficiently low temperatures in order to precipitate inhibitor substances. The manufacturing process using the former technology for the preparation of precipitates is called the “Inherent inhibitor method, Solid Solution Method or Extra high temperature slab reheating method (>1 300°C)” and referred to as “Method I” hereafter in this paper. The latter is called the “Acquired inhibitor method, Precipitation Method or Extra low temperature slab reheating method (1 100–1 200°C),” and referred to as “Method II.” Table 1 shows the classification of GO.\(^1,12-14\)

At present, all high permeability GO is manufactured using AlN as the main inhibitor.\(^7,12,15\) Therefore, the function of nitrogen (which is introduced in steel making and/or nitrided in the following operation) might be very important. Sakakura et al. reported the importance of size and distribution of AlN.\(^16\) However, although the inhibitor preparation and state of both methods are quite different mutually, the sharpness of the Goss orientation obtained by both methods is almost the same. This suggests, in general terms, that the state of AlN might not be so important, but the inhibitor substance itself might be very important.\(^12\)

In addition to these two methods, there is the possibility of the “Middle temperature slab reheating method (1 200–1 300°C),” referred to as Method III, in principle. However, few studies have been reported on this. The reason is that this method is very difficult to employ on the industrial scale, i.e., uniformity of secondary recrystallization, which depends on the uniform precipitation of inhibitors. Figure 1 shows the metallurgical concept of slab reheating on the industrial scale. To avoid the uniformity of the inhibitor-state (precipitate and solid solution), the difference in their solubility in the slab should be minimized. Therefore, the temperature of slab reheating should be extra high or extra low. Regarding such study, S. Cicale et al.\(^17\) reported a case of slab reheating at 1 300°C for the chemical composition of Method II. The metallurgical possibility has been demonstrated. However, in their study, the nitrogen content was not changed, i.e., inhibitor intensity was constant and finer precipitates could be observed.

This study aims to demonstrate the universal effect of nitriding on Al-bearing GO and the role of a combination of primary grain size, controlled with the annealing tempera-
ture, and inhibitor intensity, controlled with nitriding, in order to obtain sharp Goss secondary recrystallized texture, by the application of middle temperature slab reheating.

Meanwhile, the main factors for secondary recrystallization are inhibitor, primary grain size and primary recrystallization texture. However, as, in this study, the cold rolling reduction and other parameters for primary recrystallization texture were controlled to be the same as Method I and II, the influence of primary texture could be eliminated.

2. Experimental Procedure

Experimental conditions were controlled in order to establish the main difference as the slab reheating temperature compared with Method I. Table 2 shows the chemical composition of the specimens, which is almost same as Method I.

Experimental procedure in the laboratory for the secondary recrystallization was as follows:

Ingot was pre-rolled to 35 mm after reheating at 1100°C for 60 min. and reheated at 1250°C, at which AlN and MnS are partially precipitated. In fact, after hot rolling, 38% N as AlN, 32% S as MnS and 37% as Cu2S were precipitated.

Material was hot-rolled to a thickness of 2.25 mm. These specimens were annealed at 1200°C for 20 h at 1200°C under a 100% H2 atmosphere for purification.

As magnetic flux density is strongly connected with Goss orientation, hereafter it (B8 (T): in an applied field of 800 A/m) is used to estimate the secondary recrystallization texture. B8 was measured by the Single Sheet Tester (60 mm in width, 300 mm in length) and the average values of 8 specimens at each condition were used for analysis.

Secondary recrystallization textures were measured by the back-reflection Laue diffraction method. The cross-sectional microstructures were observed and primary grain size was measured. Precipitates of inhibitors were observed by TEM.

3. Result and Discussion

3.1 Precipitation and Primary Grain Size

As the slab reheating temperature was 1250°C, according to Iwayama et al., inhibitor substances were precipitated partially. In fact, after hot rolling, 38% N as AlN, 32% S as MnS and 37% as Cu2S were precipitated. Following the cold-rolling, the specimens were annealed in a wet atmosphere of 25% N2 and 75% H2 for decarburization and primary recrystallization. The annealing temperatures were from 825 to 875°C at every 10°C. This range of temperature is normal and is often applied to decarburization annealing of GO. Nitrogen was injected from zero up to around 0.03 mass% by changing the flow rate in the ammonia containing atmosphere and the annealing separator, mainly consisting of MgO, was coated on the specimen surfaces. The secondary recrystallization annealing was performed under a 25% N2 and 75% H2 atmosphere with a heating rate of 15°C/h up to 1200°C and maintained for 20 h at 1200°C under a 100% H2 atmosphere for purification.

Table 1. Classification of GO.

<table>
<thead>
<tr>
<th>Slab reheating temp.</th>
<th>Main inhibitors</th>
<th>Final cold rolling reduction</th>
<th>Remarks</th>
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<tbody>
<tr>
<td>Inherent inhibitor method</td>
<td>MnS</td>
<td>Light (50%)</td>
<td>Armco method</td>
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<tr>
<td>(Solid Solution Method, Extra high temperature slab reheating method) Method I</td>
<td>AlN, MnS</td>
<td>Heavy (&gt;80%)</td>
<td>Nippon Steel method</td>
</tr>
<tr>
<td>Acquired inhibitor method</td>
<td>AlN (Nitriding)</td>
<td>Heavy (&gt;80%)</td>
<td>Nippon Steel Method</td>
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<table>
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<th>Table 2. Chemical composition.</th>
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<tr>
<td>Element</td>
</tr>
<tr>
<td>C</td>
</tr>
<tr>
<td>Si</td>
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<tr>
<td>Mn</td>
</tr>
<tr>
<td>S</td>
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<tr>
<td>Cu</td>
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<tr>
<td>Al</td>
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<tr>
<td>N</td>
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<tr>
<td>Sn</td>
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87.3%.

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Figure 2 shows electron micrographs of the precipitates in the central layer after decarburization. They were primary-recrystallized at 845 and 855°C. Both fine and coarse precipitates can be observed. This feature is the state between Method I and Method II.17,25,27)

Figure 3 shows the relation between the primary annealing temperature and the primary grain diameter. This range of grain diameters is located between Method II and Method II.18,22,27,28) Furthermore, the slope against the temperature is somewhat smaller than Method II.28) This might be caused by the fine precipitation effect of partially solved inhibitors at the slab reheating stage.

3.2. Magnetic Property and Goss Orientation
3.2.1. Magnetic Property

Figure 4 shows the relation between the total N content and B8 for each primary annealing temperature.

Figure 5 shows the macrostructures after secondary recrystallization annealing. From both figures, only the no-nitriding condition gave fine grains (poor secondary recrystallization) and other conditions gave good secondary recrystallization. But some of them indicate the blunt (less sharp) Goss orientation. At around 0.015% N contents, the B8 value is somewhat high. Figure 6 shows the {100} pole figure of secondary recrystallized grains, of which B8 is 1.919T. This shows sharp Goss orientation.

Figure 7 shows the relation between the diameter, regressed by temperature, and maximum B8-values are shown in Fig. 4. As the diameter increased in size, B8 also increased and the B8 values were sufficiently high to be a
Fig. 5. Macrostructures of secondary recrystallized grains at each decaburization and annealing temperature (T), and for total Nitrogen content (N).
high permeability grade. This is the same feature as the former studies,\textsuperscript{17,22} as in the case of Method II. In their study, the total N content was around 0.02\%, which is more than this study. Furthermore, Ushigami et al. reported that the total N should be between 0.020\% and 0.029\% in Method II.\textsuperscript{25}

On the other hand, Kuroki et al. already reported that, in the case of Method I, the diameter does not change by the primary annealing temperature.\textsuperscript{27} Furthermore, they reported that the increase of the N content deteriorates the B8 value, \textit{i.e.}, no nitriding is suitable.\textsuperscript{23}

In the case of Method III, suitable total nitrogen content (0.015\%) is located between Method I (0.008\%) and Method II (0.020\% to 0.029\%), meaning the suitable inhibitor intensity in this study is located between both methods. Incidentally, inhibitor intensity depends on the chemical composition and heat treatments \textit{(i.e.}, slab heating temperature, hot rolling condition and hot band annealing condition). As mentioned above, in this study, the chemical composition is the same as Method I and only the slab reheating condition was different. Therefore, the slab reheating temperature might affect the inhibitor intensity and nitriding could compensate for it.

As shown in Fig. 3, as the temperature increased, the grain diameter generally increased. Its range is around 10 to 12 $\mu$m. Even this range of grain diameter can give sharp Goss orientation. As, in the case of Method I, where the inhibitor is finely dispersed, the primary grain diameter is somewhat small, 10 $\mu$m, and it is almost independent of the decarburization annealing temperature.\textsuperscript{27} On the other hand, in the case of Method II, the optimum primary grain diameter is around 23 $\mu$m, and it depends on the decarburization annealing temperature.\textsuperscript{22,27} Furthermore, in the case of S. Cicale, 20 $\mu$m gave the highest B8 at 1 300°C reheating. The suitable primary grain size in this study is located between Method I and Method II.

In conclusion, the two main factors (inhibitor intensity and primary grain size) of Method III for sharp Goss orientation are located between Method I and Method II. This fact strongly suggests that the combination of inhibitor intensity and primary grain size is metallurgically essential to obtain sharp Goss orientation. Figure 8 shows the relation between primary grain size and inhibitor intensity. Inhibitor intensity can be controlled with the nitriding amount in the case of Al-bearing GO in spite of the heat treatment conditions. As, of course, primary recrystallization texture is also
very important, a third axis should be added for it in Fig. 8. In the future, this effect will also be discussed.

4. Conclusion

All high permeability GO is manufactured using AlN as the main inhibitor. Two types of inhibitor preparation of GO have been realized industrially, i.e., extra high and extra low slab reheating temperature methods. In addition, from a purely metallurgical viewpoint, the possibility of a middle temperature slab reheating method was examined and the effect of nitriding was investigated. Metallurgically, this method is located between extra high and extra low temperature methods. Furthermore, it is essential that a suitable combination of inhibitor intensity and primary diameter gives sharp Goss orientation. The inhibitor intensity and primary diameter can be controlled by nitriding and heat treatment conditions (slab reheating and decarburization temperatures), respectively.

REFERENCES