Improvement of magnetic properties of an Fe-6.5 wt. % Si alloy by directional recrystallization

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We report that magnetic properties of an Fe-6.5 wt. % Si alloy can be improved through texture control by using directional recrystallization. Columnar grain structures with column sizes of $\sim 0.38 \times 1.2$ mm^2 were developed during directional recrystallization. It was found that there are low energy boundaries between columns and main textures of the specimen were {110}⟨111⟩ and {111}⟨110⟩. As a result, the coercivity of a directionally recrystallized specimen is reduced by a factor of 5 when measured along 60° away from the growth direction, as compared to a specimen consisting of ~77 μm equiaxed grains. © *2008 American Institute of Physics*.
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There has been an ever-increasing interest in obtaining desired mechanical/physical properties by controlling microstructures of materials. In the steel industry one widely used technique to change grain sizes and crystallographic textures is cold/hot rolling followed by appropriate heat treatments. Similar processes are also used to optimize physical properties of electrical steels[1] and growth directions of crystalline films.[2] Directional recrystallization is an annealing process where the sample is passed through a hot zone with a steep temperature gradient.[3] Columnar-grained structures or single crystals can be produced during the directional recrystallization by selective grain growths and competitive migrations of certain grain boundaries.[4,5] Therefore, this technique provides a useful way to change microstructure by controlling grain boundary migrations, and is especially suitable for controlling grain morphologies, crystallographic textures, and grain boundary structures.[3–7]

In this study, directional recrystallization is conducted on an Fe-6.5 wt. % Si alloy to control its textures and grain boundaries. The Fe-6.5 wt. % Si is a high-silicon material with superior magnetic properties such as high permeability, low core loss, and nearly zero magnetostriction.[8,9] Previous studies show that its magnetic properties not only depend on the silicon content but also strongly on the microstructure (e.g., grain size and texture).[10,11] However, because the ductility of those alloys decreases sharply as the silicon content increases, it is not possible to produce the Fe-6.5 wt. % Si alloy with controlled grain orientations by using traditional processes, such as cold rolling.[12] This dramatically restricts the improvement in magnetic properties of this alloy. However, as shown in this letter, the grain orientation of the Fe-6.5 wt. % Si alloy can be controlled by using directional recrystallization, and the coercivity can be dramatically reduced at a direction of 60° away from the growth direction.

An Fe-6.5 wt. % Si alloy, having compositions (in wt. %) of 0.05 C, 6.31 Si, 0.11 Mn, 0.008 P, 0.006 S, 0.005 Al, 0.04B, and the balance Fe, was first induction melted and cast in a vacuum furnace. The as-cast ingot was then homogenized and forged into a 12 mm thick plate. The forged plate was hot rolled to reduce its thickness to 2 mm and finally annealed to form a fully recrystallized structure. Starting materials for directional recrystallization were $\sim 1.5 \times 5 \times 70$ mm^3 strips, cut from the fully recrystallized plate by using electrodischarge machining (EDM). The directional recrystallization was performed in a vacuum furnace (10^{-3} Pa) with a hot zone temperature and length of 1150 °C and 5.5 mm, respectively. The temperature gradient was fixed at ~350 °C/cm and the withdrawing velocity was 3 μm/s. The experimental setup for directional annealing was described in detail in Ref 5. For comparison, isothermal annealing was also conducted in a vacuum furnace (10^{-1} Pa) at the same temperature for 30 min, which gives approximately the same annealing time as directional recrystallization.

Figure 1(a) shows a typical microstructure of the starting material, which consists of equiaxed grains with an average grain size of about 77 μm, indicating a fully recrystallized microstructure. Figure 1(b) shows its textures, which were presented as orientation distribution function (ODF) in Euler space defined by Euler angles of φ_1, Φ, and φ_2. ODF results show that crystallographic textures are scattered with weak cube, Goss, and {111} fiber textures. Figure 2(a) is a typical microstructure of an isothermally annealed (1150 °C and 30 min) specimen. As can be seen, the microstructure consists of only large equiaxed grains with grain sizes of about 350 μm, indicating a fully recrystallized and grain-grown structure. In a sharp contrast with the equiaxed grain structure of starting and isothermally annealed materials, an elongated or columnar grain structure was developed after directional recrystallization, and its typical micrograph is shown in Fig. 2(b). The grain size is measured approximately 1.2 mm at the longitudinal direction and ~0.38 mm at the transverse direction. To obtain quantitative information on the

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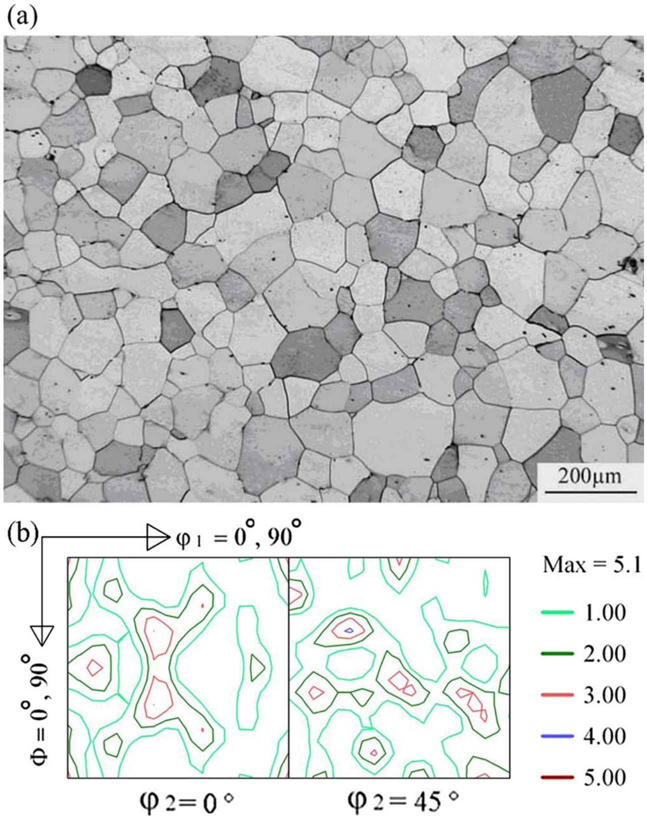

FIG. 1. (Color online) The typical (a) microstructure and (b) textures of the starting Fe-6.5 wt. % Si alloy.

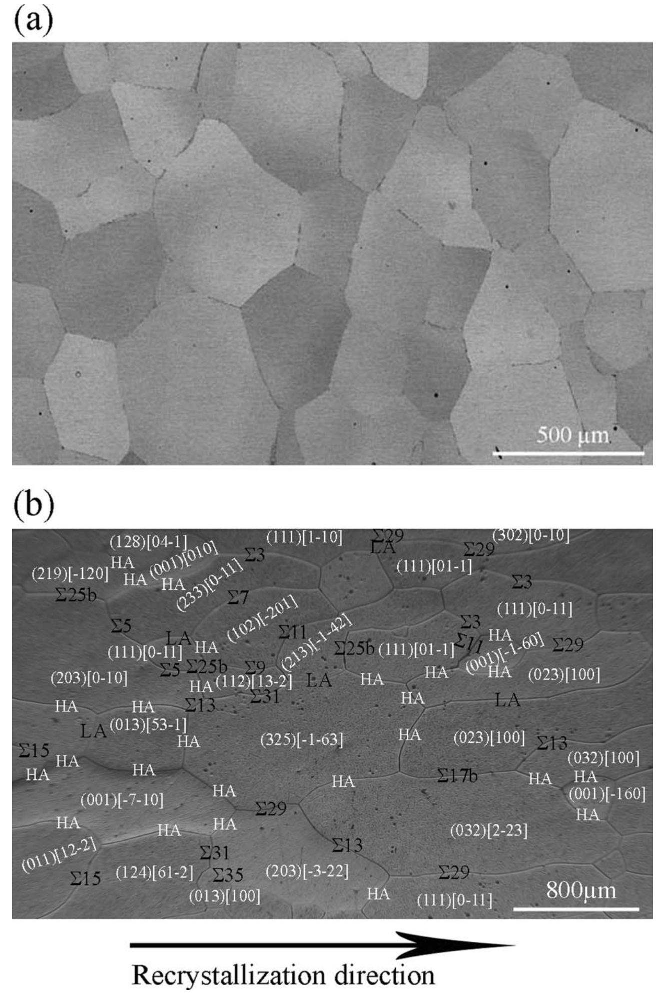

FIG. 2. Microstructures of Fe-6.5 wt. % Si alloys after (a) isothermal annealing at 1150 °C for 30 min and (b) directional recrystallization at 1150 °C. Arrow indicates the growth direction of columnar grains. LA—the low angle grain boundary and HA—the high angle grain boundary.

grain structure, measured grain orientations and calculated Σ values of the coincidence site lattice (CSL) of grain boundaries are also included in this figure.

Figure 3(a) shows the crystallographic textures in the directionally recrystallized specimen. The crystallographic textures in this specimen are significantly different from those in the starting specimen. The intensity of the {111} ⟨0*vw*⟩ fiber texture increased significantly. When cut at $\varphi_2=45°$ and $\Phi=55°$ ODF results show maxima at 0° and 60° along φ_1, corresponding to {111}⟨110⟩ components. When cut at $\varphi_2=0°$ and $\varphi_1=0°$ along Φ ODF results show maxima at 35° and 55°, corresponding to {023} ⟨100⟩ texture component. In the ODF cut at $\varphi_2=0°$ and at $\varphi_1=60°$, the orientation density along Φ also shows maxima at 35° and 55°, corresponding to {023} ⟨232⟩ texture components, which is about 10° deviation from the {110} ⟨111⟩ component. Above texture analysis indicates that main textures of directionally recrystallized specimen were {110} ⟨111⟩ and {111} ⟨110⟩.

Textures of directionally recrystallized specimen are not only different from those of the starting material, but also show difference as compared to the isothermally annealed specimen. Figure 3(b) shows representative ODF of an isothermally annealed (1150 °C and 30 min) specimen. After isothermal annealing, the intensity of the near Goss orientation increased slightly with some weakly recognizable {111} ⟨0*vw*⟩ fiber texture, which is in consistency with textures of specimens with a fully recrystallized and grain-grown microstructure.[13] However, during directional recrystallization, the microstructure was found to be a columnar-grained structure and the texture change to a {111} ⟨110⟩ dominant texture. Table I shows the main component orientations in the directionally recrystallized specimen. Misorientations among main components were characterized as low angle and CSL structures with low Σ values. It should be noted that the morphology and distribution of grains also affect the grain boundary character. To investigate the detail of misorientations among grown grains, we measured orientations and calculated Σ values of the CSL grain boundaries for the directionally recrystallized specimen, and the result is shown in Fig. 2(b). It is clear that most of boundaries between neighboring columnar grains can be characterized as low angle or CSL (with low Σ values) boundaries. It is well known that low angle grain boundaries and CSL structures with low Σ values have lower energy than random boundaries.[14] Therefore, the selective growth of columnar grains and competitive grain boundary migration appears to determine the texture evolution during directional recrystallization.

To investigate the possibility to improve magnetic properties by using directional recrystallization, coercivities of specimens were measured in a vibrating sample magnetometer (Lakeshore 7410VSM). The coercivity was measured in four directions: 0°, 45°, 60°, and 90° away from the growth direction of directional recrystallization. For comparison, coercivities of starting and isothermally annealed specimens were also measured. Specimens used for coercivity measurement are EDM-cut both before and after annealing and fol-

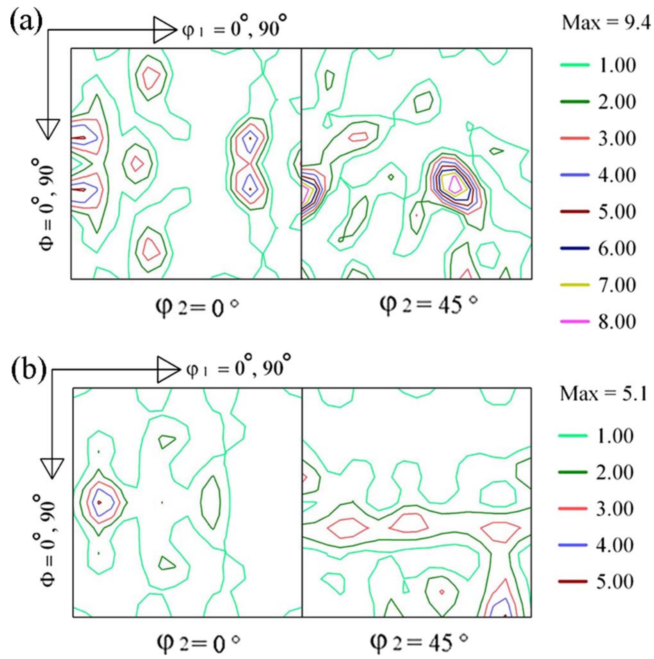

FIG. 3. (Color online) Crystallographic textures in (a) a directionally recrystallized and (b) an isothermally annealed Fe-6.5 wt. % Si specimen (φ_2=0°, 45° sections from ODF).

lowed by carefully grinding up to 1200 grit SiC paper. Final dimensions of measured specimens were $0.5\times4\times8$ mm^3. Table II lists coercivities of specimens subjected to various annealing processes. The effect of measurement direction on coercivities in both starting and isothermally annealed specimens was not pronounced. However, it is found that coercivities in the starting specimen (sample 1) were larger than those in the isothermally annealed specimen (sample 2). This is probably because the coercivity decreases as the grain size coarsens for materials with equiaxed grain structure.[15] However, for the directionally recrystallized specimen (sample 3), coercivities are dramatically different along different directions. Along 0° away from the growth direction, the coercivity increased significantly, and the value is even larger than that obtained in the starting samples with a small exquiaxed grain structure (sample 1). This probably can be explained by the effect of crystallographic textures on the coercivity. With the columnar-grained structure developing during directional recrystallization, both {110} ⟨111⟩ and {111} ⟨110⟩ components were strengthened, resulting in the increase in coercivity because both ⟨111⟩ and ⟨110⟩ are difficult directions for magnetization.[16] However, it is found that at 60° from long axis of columnar grain, the coercivity decreased to a minimum of 13.9 A/m because of the strengthening of near ⟨100⟩ and weakening of near ⟨111⟩ components in this direction. This result indicated that soft magnetic properties could be changed dramatically by directional recrystallization. For example, the coercivity has been reduced by a factor of 5 in the direction of 60° away from the growth direction for directionally recrystallized specimens, as compared to the starting material that consisted of ~77 μm equiaxed grains.

TABLE I. Misorientations of main orientation existing in directionally annealed Fe-6.5 wt. % Si samples.

φ_1, Φ, φ_2 (deg)	0, 35, 0	0, 55, 0	60, 35, 0	60, 55, 0	0, 55, 45	60, 55, 45
0, 35, 0	LA[a]	–[b]	–	–	–	–
0, 55, 0	Σ13	LA	–	–	–	–
60, 35, 0	Σ31	Σ17b	LA	–	–	–
60, 55, 0	Σ17b	Σ31	Σ13	LA	–	–
0, 55, 45	HA[c]	Σ29	Σ29	HA	LA	–
60, 55, 45	Σ29	HA	HA	Σ29	Σ3	LA

[a]LA: the low angle grain boundary.
[b]–: duplicate pairs of orientations.
[c]HA: the high angle grain boundary.

TABLE II. Coercivities of samples subjected to different annealing processing.

Angle (°)	Sample 1[a] (A/m)	Sample 2[b] (A/m)	Sample 3[c] (A/m)
0	60.0	44.4	73.4
45	71.64	39.3	31.0
60	67.6	44.5	13.9
90	47.7	28.8	28.3

[a]Starting sample with primary recrystallized grains.
[b]Sample isothermally annealed at 1150 °C for 30 min.
[c]Sample directionally annealed at 1150 °C.

In summary, directional recrystallization can be applied to change the crystallographic texture evolution and to improve the soft magnetic properties of an Fe-6.5 wt. % alloy. Our results indicate that directional recrystallization prefers the formation of low angle grain boundaries and CSL structures with low Σ values. The selective grain growth and competitive grain-boundary migration appears to control textures of the specimen by forming low energy boundaries and thus change magnetic properties.

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[1]Y. Sidor, F. Kovac, and T. Kvackaj, Acta Mater. **55**, 1711 (2007).
[2]S. J. Park, D. P. Norton, and V. Selvamanickam, Appl. Phys. Lett. **87**, 031907 (2005).
[3]M. M. Baloch and H. K. D. H. Bhadeshia, Mater. Sci. Technol. **6**, 1236 (1990).
[4]J. Li and I. Baker, Mater. Sci. Eng., A **392**, 8 (2005).
[5]Z. W. Zhang, G. L. Chen, and G. Chen, Mater. Sci. Eng., A **422**, 241 (2006).
[6]Z. W. Zhang, G. L. Chen, and G. Chen, Mater. Sci. Eng., A **435**, 573 (2006).
[7]Z. W. Zhang, G. Chen, and G. L. Chen, Acta Mater. **55**, 5988 (2007).
[8]H. Honma, Y. Ushigami, and Y. Suga, J. Appl. Phys. **70**, 6259 (1991).
[9]H. Ninomiya, Y. Tanaka, A. Hiura, and Y. Takada, J. Appl. Phys. **69**, 5358 (1991).
[10]K. Moras and W. Riehemann, J. Magn. Magn. Mater. **254**, 29 (2003).
[11]R. E. Lenhart, J. Appl. Phys. **35**, 861 (1964).
[12]M. Komatsubara, K. Sadahiro, O. Kondo, T. Takamiya, and A. Honda, J. Magn. Magn. Mater. **242**, 212 (2002).
[13]T. Ros-Yanez, Y. Houbaert, O. Fischer, and J. Schneider, J. Mater. Process. Technol. **143**, 916 (2003).
[14]T. Watanabe, Scr. Metall. **27**, 1479 (1992).
[15]G. Herzer, Scr. Metall. **33**, 1741 (1995).
[16]J. S. Kouvel and R. H. Wilson, J. Appl. Phys. **32**, S276 (1961).