Secondary recrystallization induced by composite precipitates in magnetostrictive Fe-Ga alloy thin sheet

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Abstract. Secondary recrystallization of Goss ([110]<001>) texture is successfully produced by using nanometer-sized composite inhibitor in primary cold rolled Fe 81Ga19 alloy sheet. Nanometer-sized sulfides consisting of MnS and Cu 2S are dispersedly precipitated during hot rolling and normalization as inhibitors for secondary recrystallization. Strong γ texture with peak at {111}<112> and weak Goss texture distributed through the sheet thickness in primarily annealed sheets can provide favorable environment for the secondary recrystallization. The higher onset and finishing temperatures of secondary recrystallization confirm the stronger inhibition and driving force originated from the composite inhibitors. Centimeter-sized Goss grains and large magnetostriction coefficient are obtained by primary cold rolling methods without surface energy effect. The result indicates that complete secondary recrystallization in Fe-Ga alloy can be realized by precisely controlling inhibitor characteristic and primary recrystallization texture.

1. Introduction

Fe-Ga alloy is an advanced magnetostrictive material for actuators and sensors in terms of combining excellent mechanical and magnetostrictive properties [1-3]. It is desirable to produce η (<100>⁄RD) textured Fe-Ga sheets due to the largest magnetostriction coefficient along <100> direction and lower eddy current loss in high frequency use. A series of work has been conducted to promote η texture in Fe-Ga sheet through secondary recrystallization by conventional rolling and annealing process [4-7]. According to current knowledge, secondary recrystallization of η texture requires two necessary conditions: (1) enough pinning force for matrix grains normally originating from precipitate particles (called as inhibitors); (2) preferred growth for η grains by grain boundary mobility advantage or additional driving force from surface energy difference or gradually weaken inhibition.

It is concluded that current methods to promote secondary recrystallization in Fe-Ga alloy mainly include: (a) NbC particles with size of 1-5 μm were added to the molten Fe81Ga19 alloy to inhibit the normal grain growth (NGG) [4,8], while the abnormal grain growth (AGG) of Goss ([110]<001>) texture was induced by surface energy effect from H2S atmosphere [5]. Due to the weak stability and pinning of the coarse NbC particles, complete secondary recrystallization has been achieved by addition of 1-2.5 at.% NbC particles combined with the high temperature annealing of 1200-1250 °C for 2-4 hours [9]. (b) Micronmeter- or nanometer-sized NbC particles were precipitated in rolling and annealing process of Fe81Ga17 and Fe82Ga4.5Al13.5 alloy sheets, the AGG of Goss texture was induced by surface energy effect under sulfur atmosphere combined with strengthen of Goss texture in primary recrystallization prepared by initial sharp <100>-oriented columnar grains and intermediate annealing [6,10,11]. So far, strong Goss and large magnetostriction coefficient has been achieved by secondary
recrystallization with the combination of inhibitor and surface energy modification during high temperature annealing. However, secondary recrystallization texture in Fe-Ga sheets has not been well regulated so that the costly application of surface energy effect and unconventional processing method are necessary. In our previous work [12], nanometer-sized sulfides were dispersedly precipitated during hot rolling, the AGG of Goss texture was produced well-matched pinning effect and driving force by precise control of inhibitor and primary recrystallization texture in two-stage cold rolled Fe$_81$Ga$_{19}$ alloy sheets.

In the grain oriented silicon steel, a few number of Goss grains in primary recrystallization are obtained by primary cold rolling with a large reduction ratio, while the nanometer-sized inhibitors, consisting of MnS, Cu$_2$S, AlN particles, mainly precipitate at hot rolling and normalization, which provides strong driving force for secondary recrystallization. Finally, sharp Goss texture with lower deviation is produced by prefect-matched pinning force and driving force of secondary recrystallization [13-15]. Primary cold rolling with large reduction ratio significantly reduces the quantity and deviation of Goss nuclei in primary recrystallization, and then provides sufficient growth space for the secondary recrystallization of exact Goss texture. However, it requires stronger inhibition accompanied with more precise matching of precipitation regression process with intrinsic preferred mobility of Goss grain in higher secondary recrystallization temperature. In this paper, nanometer-sized composite inhibitor was precipitated to induce secondary recrystallization of Goss texture in Fe$_81$Ga$_{19}$ alloy sheet prepared by primary cold rolling with a large reduction ratio, the relationship between inhibitor characteristics and secondary recrystallization was studied.

2. Material and methods
The Fe$_81$Ga$_{19}$ alloy ingots with nominal chemical composition (in weight %) of 0.03% C, 22.65 % Ga, 0.25% Mn, 0.15% Cu, 0.01% S and balance Fe were prepared from pure Fe (99.9%) and Ga (99.99%), pure Cu (99.99%) and master alloy of Fe-C and Fe-Mn by induction melting. The ingots were hot rolled to 2.3 mm at 1200 °C with finishing temperature of 850 °C. Afterwards, hot bands were normalized annealed at 1000°C and then cold rolled to 0.45 mm at 200 °C with 80% reduction. The cold rolled sheets were annealed at 850 °C for 10 min in a wet atmosphere of 75% H$_2$ and 25% N$_2$ for primary recrystallization, then heated to 1100 °C at a rate of 20 °C/h in an atmosphere of 75% N$_2$ and 25% H$_2$, and finally annealed at 1200 °C for 1 h in 100% H$_2$ for purification.

The orientation distribution functions (ODFs) of the specimens were measured and calculated at different thickness layers based on X-ray diffraction. The measured layer is defined as the parameter S=2ad, where a and d are the distances from the center and sheet thickness, respectively. Electron backscattered diffraction (EBSD) analysis was also performed to investigate the texture evolution of rolling and annealing process. The precipitates were prepared by carbon extraction-replica technique and studied via transmission electron microscope (TEM) equipped with an energy dispersive X-ray spectroscopy (EDS). Magnetostriiction values under applied field from 0 to ± 3000 Oe were measured by strain gauges with gauge area of 4 mm × 6 mm (base area of 6.3 mm × 10.5 mm) positioned along the rolling direction. The saturated magnetostriiction coefficient was calculated by $(3/2)\lambda_s=\lambda_{\parallel}\lambda_{\perp}$, where $\lambda_{\parallel}$ and $\lambda_{\perp}$ are the maximum magnetostriiction coefficients under the magnetic field applied parallel and perpendicular to the rolling direction (RD), respectively.

3. Results and Discussion
Figure 1 shows the feature of precipitates in normalized bands and primary recrystallized sheets. The precipitates are determined as MnS and Cu$_2$S by EDS analysis. A number of spherical, rectangular and lamellar precipitates with size of 10–30 nm are observed in normalized bands. After cold rolling and primary recrystallization annealing, fine precipitates of MnS and Cu$_2$S are still dispersed in the matrix with size of 20–60 nm, which is critical for efficient pinning effect on primary recrystallized grains [13-15]. The nanometer-sized precipitates can be attributed to the large nucleation rate due to high density of dislocations as preferred nucleation sites by heavy hot rolling and subsequent rapid cooling together with the regulated solubility products of MnS and Cu$_2$S precipitates by composition design [14,16].
Figure 1. TEM micrograph and EDS of precipitates in (a) normalized band and (b) primary recrystallized Fe$_{81}$Ga$_{19}$ thin sheet.

Figure 2 shows microstructure and texture of hot-rolled, cold rolled and primarily recrystallized sheets. The hot rolling microstructure and texture (figure 2a,b) exhibit a through-thickness gradient, where fragmented grains oriented by strong Goss and weak brass (⟨110⟩⟨112⟩) texture occupy subsurface layer and (S=0.5) and severely elongated grains with strong α fiber (⟨110⟩//RD) with peak at {001}⟨110⟩ and weak γ fiber (⟨111⟩//ND) distributed in center layer (S=0). The microstructure of cold rolled sheet is composed of elongated grains with less in-grain shear bands through sheet thickness (figure 2c). The first cold rolling texture is characterized by strong α fiber with peak at {112}⟨110⟩ and γ fiber (⟨111⟩//ND) with peak at {111}⟨110⟩ at both layers (figure 2d), which is similar to the rolling textures in electrical and ultra-low-carbon steel[17-19]. The primary recrystallized microstructure consists of nearly homogeneous grains (18~21 μm) through thickness. The primary recrystallization texture is characterized by strong γ fiber with a peak at {111}⟨112⟩ and weak Goss, distinct from the reported primary recrystallization texture in Fe-Ga sheet [11,20,21]. Strong {111}⟨112⟩ can promote the abnormal growth of secondary recrystallized Goss grains in terms of high grain boundary mobility as in grain-oriented silicon steel [22-24].
Figure 3 shows microstructure evolution during high temperature annealing by interrupted experiment method. Average grain size increases slowly from 20 μm at 800 °C to 26 μm at 950 °C, indicating that normal grain growth is effectively inhibited by nanometer-sized precipitates. Secondary recrystallization occurs at 1000 °C and nearly completes at 1100 °C. This temperature range is much higher and narrower than Fe-Ga-NbC sheet with the application of surface energy [6,12]. Average grain size of matrix grains further increases from 31 μm to 43 μm as temperature arise from 1000 °C to 1050 °C (figure 4), which means the pinning gradually decrease due to the coarsen of nanometer-sized precipitates as temperature exceeds 1000 °C.

According to EBSD analysis on interrupted samples (figure 5), the abnormally grown grains are identified as Goss orientation by EBSD. The deviation angle increases from 5~8° at 1000 °C with grain size of ~3 mm to 8~15° at 1200 °C with grain size of ~10 mm. This result suggests that the complete secondary recrystallization of sharp Goss texture can be induced in primary cold rolled Fe-Ga sheets by controlled precipitation of nano-sized inhibitor without the application of surface energy effect.

The magnetostriction coefficient of annealed specimens interrupted at different temperatures is shown in figure 6. The occurrence of abnormal grain growth near 1000 °C leads to an evidently increased magnetostriction coefficient of 95±19 ppm relative to 69±6 ppm at 950 °C. The magnetostriction coefficient reaches up to 164±21 ppm when centimeter-sized Goss grains develop at 1000 °C. The magnetostriction coefficient as high as 210±25 ppm is further obtained after annealing at 1200 °C, corresponding to complete secondary recrystallization of Goss grains. This value is much larger than those reported on Fe-Ga alloys with the help of micrometer-sized inhibitors [4,10], but lower than the cases with both inhibitors and surface energy [5,6,9]. This is attributed to the larger deviation angle after secondary recrystallization.
Figure 5. Orientation image maps of Fe$_{81}$Ga$_{19}$ thin sheet extracted at (a) 950 °C, (b) 1000 °C, (c) 1050 °C and (d) 1200 °C.

The secondary recrystallization within a relatively higher and narrower temperature range indicates that the selected inhibitor and its morphology during hot rolling and normalization can be used for the secondary recrystallization of Goss texture in Fe$_{81}$Ga$_{19}$ alloy. However, large deviation of Goss texture after secondary recrystallization implies the inhibitor force and its thermal stability of the inhibitor system cannot match well with the AGG of accurate Goss grains. The rapidly coarsening of the nanometer-sized sulfides at annealing temperature higher than 1000°C reduces the driving force of preferred growth of accurate Goss grains. Accordingly, it is concluded that the critical requirements for secondary recrystallization process without the application of surface energy effect in Fe-Ga alloy includes stronger inhibition to primary grains, gradually dissipated inhibition force and precise matching with intrinsic preferred mobility of Goss grain during high temperature annealing.

Figure 6. Variation of magnetostriction coefficients (3/2$\lambda_s$) with magnetic field and annealing temperature of Fe$_{81}$Ga$_{19}$ thin sheet.

4. Conclusion
In the present study, the composite inhibitor composed of nanometer-sized sulfide particles is dispersedly precipitated in hot rolling and normalization annealing. The strong inhibiting force during high temperature annealing promotes Goss grains to grow abnormally by consuming primary
recrystallized matrix grains. Centimeter-sized secondary recrystallization Goss grains and large magnetostriction coefficient as high as 235 ppm is obtained without surface energy effect in primary cold rolled Fe$_{81}$Ga$_{19}$ alloy sheet. The result indicates that complete secondary recrystallization can be realized based on the precise design and control of inhibitor and primary recrystallization microstructure/texture in Fe-Ga alloy by conventional rolling and annealing methods.

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