# Evolution of Composite Nanometer-sized Inhibitors During Secondary Recrystallization in $Fe_{81}Ga_{19}$ Sheet

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**Keywords:** Inhibitor, goss, particle pinning, $Fe_{81}Ga_{19}$ alloy.

**Abstract:** A large number of fine composite nanometer-sized inhibitors are obtained in $Fe_{81}Ga_{19}$ alloy sheet by precipitating during hot rolling. The inhibitor particle sizes distributed in primary recrystallization sheet are mainly in the range of 20-60 nm. Composite nanometer-sized precipitates (MnS and $Cu_2S$) as inhibitors of secondary recrystallization can effectively pin the matrix grains to grow up. During secondary recrystallization annealing, centimeter-sized Goss grains are produced by precisely controlling the speed of the coarsening of precipitates and intrinsic mobility of Goss grains. Finally, the magnetostriction coefficient up to 265 ppm is obtained.

## 1. Introduction

Fe-Ga alloy (Galfenol), as a new type of magnetostrictive material, has low saturation magnetic field, high tensile strength, and good temperature feature. It can be widely applied in sensors, actuators, and energy harvesting equipments[1, 2]. However, the magnetostrictive properties of Fe-Ga alloys have significant anisotropy. The magnetostriction coefficient is as high as 400 ppm along the <100> direction, and is only 40 ppm along the <111> direction. In addition, Fe-Ga alloys have high electrical conductivity and are liable to cause severe eddy current losses when used in high-frequency alternating fields. Therefore, it is desirable to produce Fe-Ga alloy sheets with sharp η texture (<100>//RD). Secondary recrystallization (also known as the abnormal growth of grains (AGG) of Goss grains is an effective way to obtain the <100> orientation in Fe-Ga alloy sheets[3-7]. The effective pinning force by second phase particles distributed in the $Fe_{81}Ga_{19}$ sheets is essential to inhibit normal grain growth (NGG) and lead to AGG. Na et al introduced NbC particles with diameter of 1~5 μm as an inhibitor in $Fe_{0.81}Ga_{0.19}$ plus 1mol.%NbC sheets. In the stage of annealing at 1200 °C, the surface energy of $H_2S$ atmosphere was used to induce the abnormal growth of Goss grains, finally, the magnetostriction coefficient as high as 292 ppm was obtained[8, 9]. Yuan et al proposed that micrometer-sized NbC particles were also an effective way for inducing Goss grains in $Fe_{83}Ga_{17}$ alloy sheets with the help of surface energy under sulfur atmosphere, and obtained the centimeters-sized Goss grains with a magnetostriction coefficient of ~245 ppm[6, 10]. The triggering mechanism of the secondary recrystallization during the annealing process mainly depends on the difference of grain boundary energy between Goss grains and non-Goss grains. High energy Goss grain boundaries have high mobility and are more easily depinned than other grains during continuous heating process.

Once depinned, Goss grains are expected to grow as fast as possible, which induce the abnormal growth of Goss grains[11].

However, the surface energy effect can be used to achieve the sharp Goss texture during secondary recrystallization annealing stage of the Fe-Ga alloy sheets, which also shows that there are still some problems in the composition and control of the inhibitor system. In this paper, the composite nanometer-sized sulfides MnS and $Cu_2S$ were choosed as inhibitors. A matching of the inhibitor coarsening rate and the growth rate of secondary recrystallization Goss grains was achieved, and finally a sharp secondary recrystallization Goss texture was formed in the Fe-Ga alloy sheets.

## 2. Experimental Procedures

The components of $Fe_{81}Ga_{19}$ alloy are (in weight%) 22.65% Ga, 0.01% C, 0.20% Mn, 0.10% Cu, 0.02% S and balance Fe were prepared from pure Fe, Ga and Cu elements and Fe-C and Fe-Mn alloys. The raw materials were melted in a non-consumable vacuum arc furnace. After being melted, electromagnetic stirring was performed. The melting and stirring were repeated several times to ensure the uniformity of the composition. The ingots were heated at 1200 °C for 30 min, and hot rolled to 2.2 mm with finishing temperature of 800 °C. Subsequently, the hot-rolled sheets were dwelled at 1000 °C for 5 min, followed by air-cooling to 900 °C and quenched with boiled water, then cold rolled to 0.3 mm. By controlling the $H_2$ and $N_2$ flow rates with a ratio of 3:1 in a wet atmosphere, the cold rolled sheets were annealed at 800 °C for 5 min to achieve decarburization and complete the primary recrystallization. Then the sheets were heated to 1050 °C at a rate of 15 °C/h with a flow ratio of $H_2$ and $N_2$ of 1:3 and finally annealed at 1200 °C for 2 h in a pure $H_2$ atmosphere.

The micro-texture was investigated in a field emission gun scanning electron microscope (SEM, JEOL6500F) with an electron backscatter diffraction (EBSD) acquisition system. The orientation distribution functions (ODFs) based on EBSD by Bunge notation. The precipitates microscope (HRTEM, JEM2100F) equipped with an energy dispersive X-ray spectroscopy (EDS). Magnetostriction values at applied field from 0 to ± 1200 Oe were measured by strain gauge positioned along rolling direction. The saturated magnetostriction coefficient was calculated by $(3/2)\lambda_S = \lambda_{\parallel} - \lambda_{\perp}$ where $\lambda_{\parallel}$ and $\lambda_{\perp}$ are the maximum magnetostriction coefficients under the magnetic field applied parallel and perpendicular to rolling direction (RD), respectively[11, 12].

## 3. Results and Discussion

After hot rolling, boiled water quenching, appropriate cold rolling and primary recrystallization annealing, the second-phase particles distributed dispersedly were precipitated in $Fe_{81}Ga_{19}$ sheets, and the characteristics of the precipitates are shown in Figure 1 and Figure 2. The precipitates were determined to be MnS and $Cu_2S$ by EDS analysis. The fine spherical and rectangular precipitates of MnS and $Cu_2S$ with size of 20-60 nm were dispersed in the matrix.

The high density of dislocations, (which can be used as preferential nucleation sites for the second-phase particles), formed by $Fe_{81}Ga_{19}$ sheets during hot rolling and subsequent rapid cooling is the main reason for the formation of nanometer-sized precipitates.

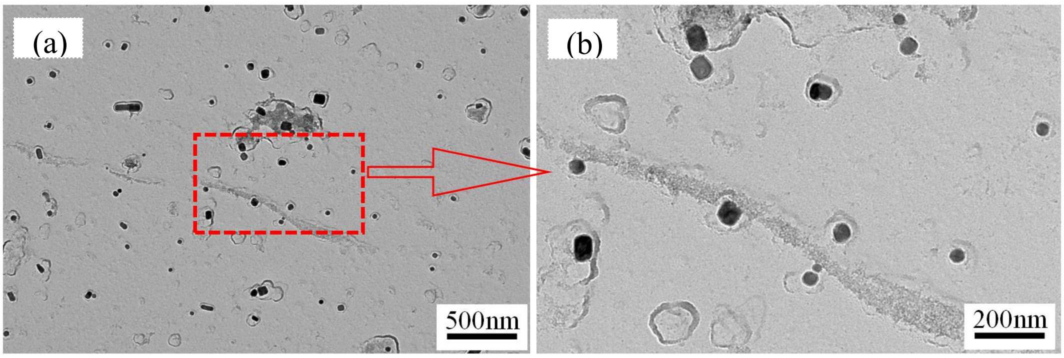


Figure 1: Characteristics of the precipitation phase in primary recrystallization sheets.

It is well known that the pinning force that inhibit grain boundary movement introduced by the second-phase particles is called the Smith-Zenner pinning force. It is expressed as $F_Z = (3/2)cf_v r^{-1}$, where c is the grain boundary energy, $f_v$ is the volume fraction of the particles, and r is the average radius of the second-phase particles. According to the formula, the smaller the particle size of the second-phase particles, the larger the volume fraction, the pinning effect is greater. In this paper, the second-phase particles are used to pin the matrix grains, and take advantage of the difference in the grain boundary mobility between Goss grains and non-Goss grains to achieve the abnormal growth of Goss grains, similar to grain-oriented silicon steel[13, 14].

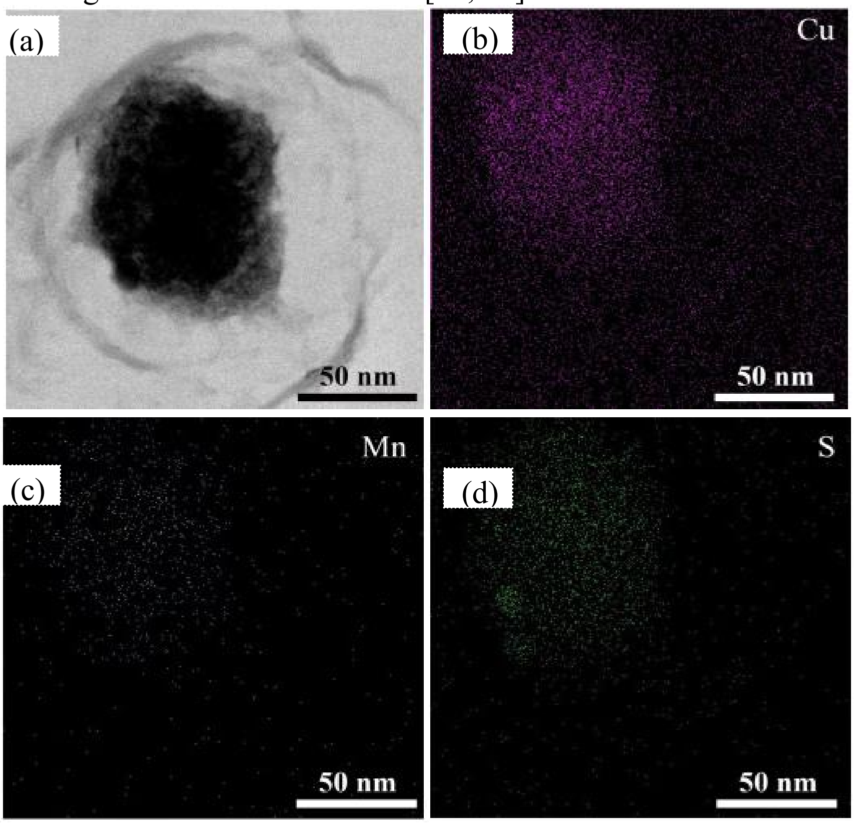


Figure 2: Energy spectrum characteristics of the precipitation phase in primary recrystallization sheets（a）TEM;（b）Cu;（c）Mn;（d）S.

The evolution of the microstructure during secondary recrystallization annealing by interrupted annealing is shown in Figure 3. When the temperature rose to 950 °C, the local grain size was significantly higher than the matrix grain size, and secondary recrystallization began to occur.

However, the matrix grain size was ~22 μm, which shows that the nanometer-sized precipitates effectively inhibit the growth of matrix grains. The magnetostriction coefficient was only 52±8 ppm (Figure 4).

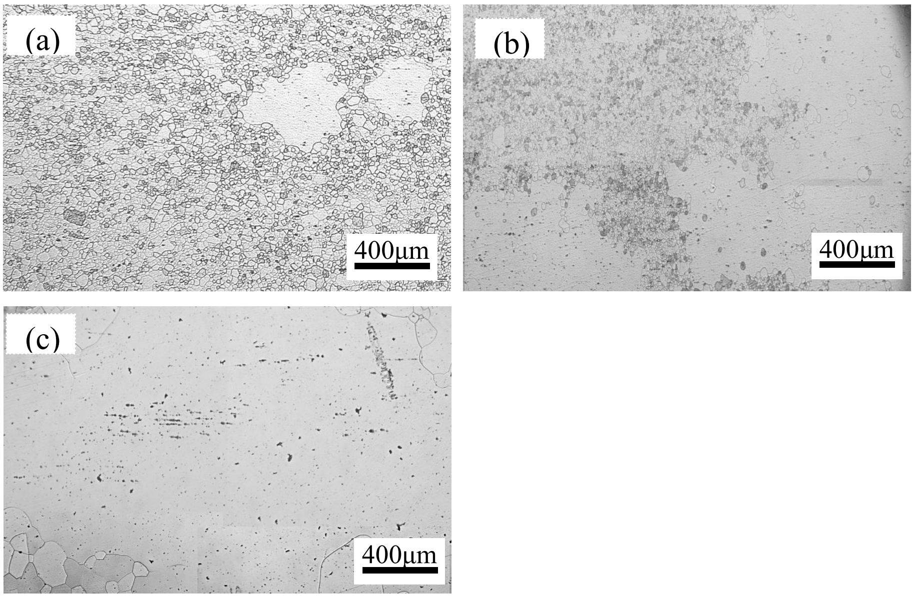


Figure 3: Microstructure evolution of Fe81Ga19 sheets during secondary recrystallization annealing (a) 950 °C (b) 1000 °C (c) 1200 °C.

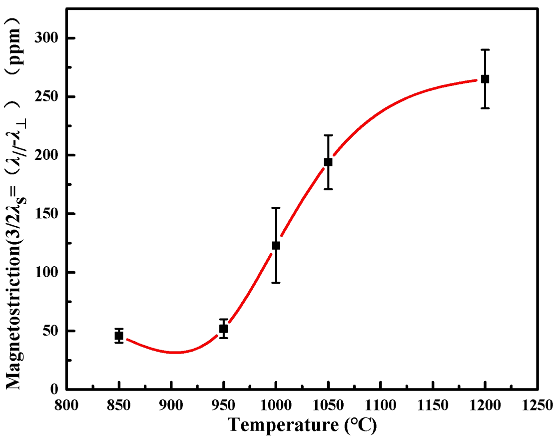


Figure 4: Variation of magnetostriction coefficients with annealing temperature of $Fe_{81}Ga_{19}$ sheets.

With the increase of the annealing temperature, the abnormal grown grains gradually swallowed up the matrix grains, and the grain size reached ~3 mm at 1000 °C. But the matrix grains did not grow up significantly which the size was ~32 μm. The magnetostriction coefficient increased from 52±8 ppm at 950 °C to 123±32 ppm at 1000 °C. After annealing at 1200 °C, the abnormal grown grains occupied about 95% of the sample surface area, only individual grains that were not swallowed up remained at the edge of the sheets. The abnormal grown grain size reached the centimeter-size, and finally a $Fe_{81}Ga_{19}$ sheets with a magnetostriction coefficient of up to 265±25 ppm was obtained.

Figure 5 shows the orientation image maps (OIM) and ODF constant $\varphi_2$ cross-section of the samples at 1200 °C. It can be seen that the abnormal grown grains are Goss. The magnetostriction coefficients strongly depend on the crystalline orientation. This result shows that, by controlling the precipitation of nanometer-sized inhibitors without applying surface energy, a sharp secondary recrystallization texture is eventually formed.

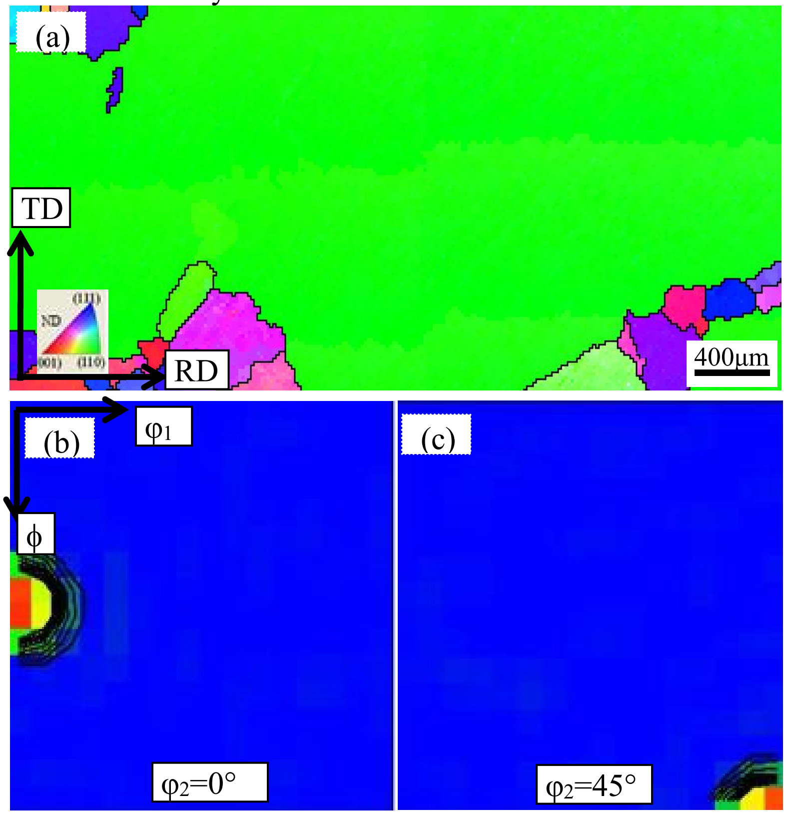


Figure 5: Orientation image maps (OIM) and ODF ((b) φ2=0°(c) φ2=45°) of $Fe_{81}Ga_{19}$ sheets at 1200 °C.

To help in understanding the AGG of Goss grains and the evolution of inhibitor particles during the secondary recrystallization annealing, several $Fe_{81}Ga_{19}$ samples extracted at different temperatures were investigated. Figure 6 shows the orientation image maps (OIM) of samples extracted at 900 °C, 950 °C, 1000 °C and TEM micrographs of the precipitation at the corresponding temperature.

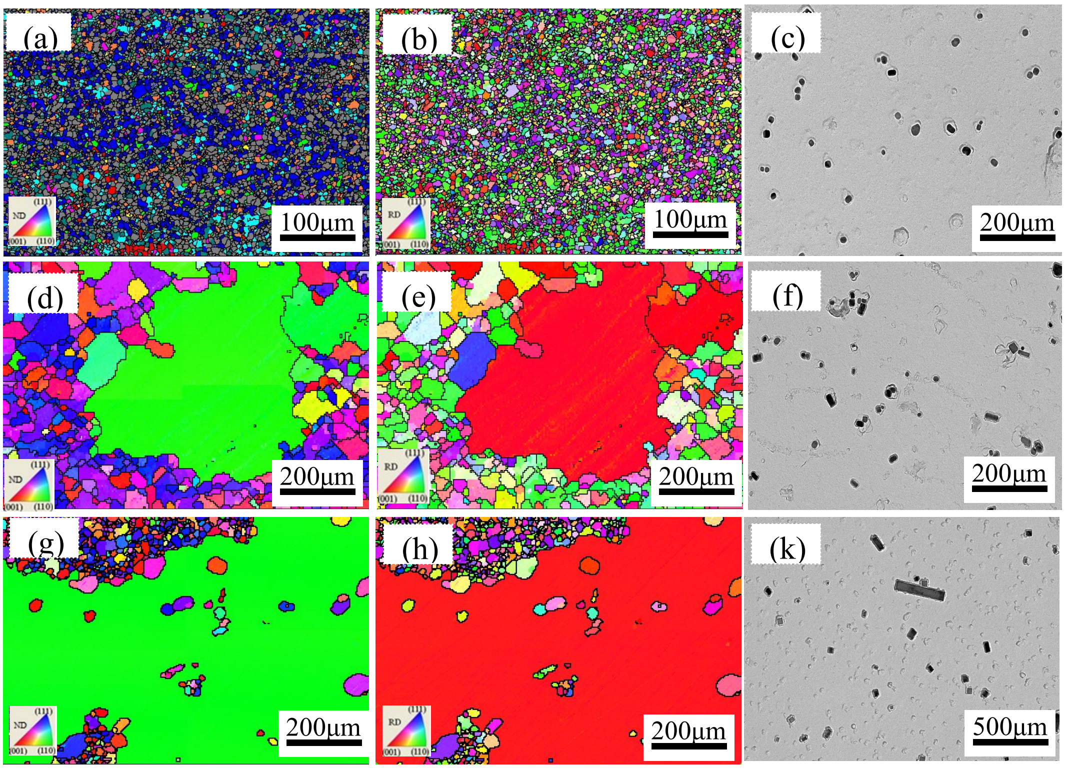

Figure 6: Orientation image maps of Fe81Ga19 sheets and TEM micrographs of inhibitor particles (a~c) 900 °C (d~f) 950 °C (g~k) 1000 °C.

It can be seen that NGG was strongly suppressed before 900 °C, the average grain size was only ~18 μm in diameter at 900 °C, which uniformly distributed in the matrix without occurring AGG of Goss grains. The textures in Figure 6 (a) and (b) were dominated by strong γ-fiber and weak Goss texture, which played a vital role in AGG of Goss grains. Lots of grain boundaries in γ deformed grains act as nucleation sites for {111}<112> recrystallized grains[15, 16], while the strong γ-fiber can promote the abnormal growth of secondary recrystallized Goss grains in terms of high grain boundary mobility as in grain-oriented silicon steel[17-19]. Therefore, the appropriate intensity of γ texture in the matrix provides a good growth environment for the abnormal growth of Goss grains. Meanwhile, the size of the precipitated particles dispersed in the matrix was mainly distributed in the range of 20-60 nm, which had a strong pinning force and a good effecton inhibiting the growth of the matrix grains, and the distribution density reached $23.4\times10^8/cm^2$, as shown in Figure 7 (a) and (b). As the temperature increased to 950 °C, the matrix grains with size of ~22 μm in diameter were still effectively pinned, but the abnormal growth of Goss grains occurred, as shown in Figure 6 (d) and (e). The inhibitor particles increased from 20-60 nm at 900 °C to 40-80 nm at 950 °C respectively. At 1000 °C, some island-like grains inside the Goss grains were observed. The matrix grain size was about 32 μm, the size distribution of inhibitor particles was in the range of 60-120 nm. It shows that the coarsened precipitates lead to a decreasing pinning force which lose their pinning effectiveness to Goss grains, and induce to AGG of Goss grains. Once depinned, Goss grains are expected to grow as fast as possible to acquire large size advantage and complete AGG. Finally, Goss grains relying on the advantage of size and grain boundary energy swallowed up the matrix grains and occupied 95% of the area in $Fe_{81}Ga_{19}$ sheets, forming a sharp Goss texture.

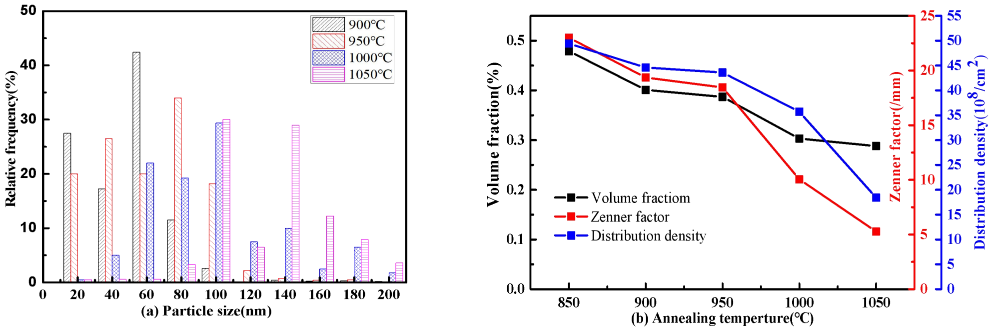


Figure 7: Variation of inhibitors in $Fe_{81}Ga_{19}$ sheets during secondary recrystallization annealing (a) size, (b) volume fraction, distribution density and pinning force.

## 4. Conclusion

A large number of fine composite nanometer-sized inhibitors dispersed in the matrix were obtained in $Fe_{81}Ga_{19}$ sheets by a rolling process in which the temperature was maintained at 1000 °C for 5 min, and then air-cooling to 900 °C to boiled water quenching, followed by cold rolling at 400 °C. After the primary recrystallization, the inhibitor particle size was mainly distributed in the range of 20-60 nm. Composite nanometer-sized inhibitors (MnS and $Cu_2S$) as inhibitors of secondary recrystallization could effectively pin the growth of matrix grains. During secondary recrystallization annealing, it gradually coarsens to achieve a precise matching with intrinsic mobility of Goss grains, and finally induces secondary recrystallization of Goss grains in $Fe_{81}Ga_{19}$ alloy sheets.

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