

Effect of Two-Step Annealing on Remanence of Fe-Pt Films Electroplated on Cu Substrates

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We have already reported a fabrication method of Fe-Pt thick film magnets using electroplating and good film's hard magnetic properties. The present study investigates two-step-annealing, consisting of low-temperature annealing (LT-annealing, 300°C) and high-temperature one (HT-annealing, 700°C) for ordering the Fe-Pt crystalline phase. We electroplated Fe-Pt films on a Cu sheet and carried out the two-step-annealing for the films. As a result, the remanence of the two-step-annealed Fe-Pt films varied depending on the holding time of the LT-annealing, and we obtained a high remanence at the holding time of around 10 min. As a next step, we annealed the Cu sheets at 300°C and electroplated Fe-Pt films on the 300°C-annealed Cu sheets. In this case, no noticeable variation in remanence was observed. As the structural change of the Cu sheet by the 300°C-annealing was confirmed using X-ray analysis, the structural change of the Cu substrate is a factor for the remanence variation by the LT-annealing. In other words, we found that the remanence can be controlled by using the structural changes of the Cu substrate.

Keywords : electroplating, Fe-Pt alloys, annealing, hard magnetic materials, magnetic properties

1. Introduction

L1₀-ordered TM-Pt films (TM: Fe, Co) have good hard magnetic properties, and they are attractive small-sized magnets for medical devices because of their good biocompatibility and high corrosion resistance [1, 2]. Owing to their high remanence and coercivity, TM-Pt alloys have been investigated for use as small magnets of MEMS (Micro Electro Mechanical Systems) applications [3, 4].

Chemical processes such as electroplating and electroless plating are generally preferred over physical ones such as sputtering and vapor deposition due to the high economic viability of the fabrication process, and some researchers reported electroplated TM-Pt thick films with high coercivity [5-8]. We also reported the fabrication process of electroplated Fe-Pt films and investigated

electroplating processes and structural and magnetic properties of the films [9].

Our previous studies revealed that the coercivity of Fe-Pt films depends on the Na and Cl ion concentrations in the plating bath, indicating that electroplating conditions significantly influence the films' magnetic properties [10-12].

Typical Fe-Pt films prepared by electroplating methods have the fcc (face-centered cubic, A1) structure and low coercivity. We need high-temperature annealing to transform the crystalline structure from fcc to fct (face-centered tetragonal, L1₀) structures to obtain high coercivity. The present study focuses on the annealing process for L1₀ ordering of the fcc Fe-Pt crystalline phase and employs two-step-annealing, consisting of low-temperature annealing (LT-annealing, 300°C) and high-temperature one (HT-annealing, 700°C). Our previous study on electroplated Fe-Co films [13] showed that 300°C-annealing causes the structural change of the Fe-Co crystalline phase around the Cu substrate and improves the films' magnetic properties. To improve the hard magnetic properties of the Fe-Pt films through LT-annealing, we investigated the effect of two-step-annealing on the coercivity and remanence values of the films.

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2. Experimental Procedures

2.1. Electroplating

We electroplated 10 μm -thick $\text{Fe}_{50}\text{Pt}_{50}$ films on Cu substrates (0.5-mm-thick Cu sheet, Nilaco Corporation). Table 1 shows bath conditions. Citric acid and $\text{NH}_4\text{OSO}_2\text{NH}_2$ were added to the baths to suppress the oxidation of Fe ions during plating and to improve the solubility of the Pt reagent, respectively. Table 2 shows the plating conditions. The conditions were the same as those for our previous study [14].

2.2. Annealing

Figure 1 shows the annealing pattern used in this study. Since as-plated Fe-Pt films have an fcc structure and show low coercivity, we need high temperature annealing (described as HT-A) to transform the film's structure from the fcc structure to the fct one. Our previous studies employed 700°C for five minutes in vacuum as HT-A

Table 1. Bath conditions.

Reagents	Contents
$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$	5 g/L
$\text{Pt}(\text{NH}_3)_2(\text{NO}_2)_2$	10 g/L
Citric acid($\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$)	30 g/L
$\text{NH}_4\text{OSO}_2\text{NH}_2$	20 g/L

Table 2. Electroplating conditions.

Conditions	Values
Bath temperature	70°C
Anode	Pt mesh
Cathode (Substrate)	Cu sheet (0.5 mm in thick)
Plating area	5 mm $\times$ 5 mm
Plating time	15 min

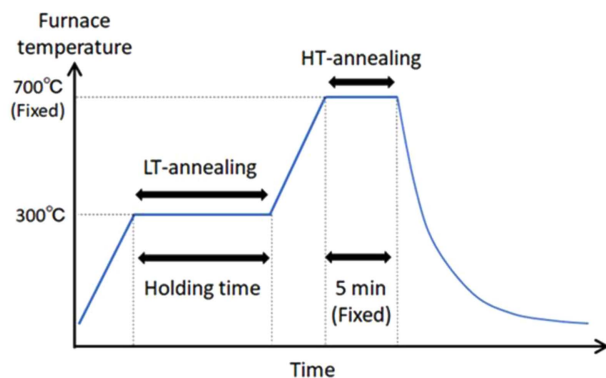


Fig. 1. (Color online) Annealing pattern.

Table 3. HT-ANNEALING CONDITIONS.

Conditions	Values
Temperature	700°C
Heating rate	100°C/min
Holding time	5 min
Atmosphere	Vacuum (3.0–4.0 $\times 10^{-3}$ Pa)

Table 4. LT-ANNEALING CONDITIONS.

Conditions	Values
Temperature	300°C
Heating rate	100°C/min
Holding time	3–20 min
Atmosphere	Vacuum (3.0–4.0 $\times 10^{-3}$ Pa)

conditions, and the same conditions were used (Table 3). The present study inserted low temperature annealing (described as LT-A) before the HT-A, as shown in Fig. 1, referred to as “two-step-annealing”. Table 4 shows the LT-A conditions. The LT-A temperature was set at 300°C, and the holding time varied from 3 to 20 min.

2.3. Measurements

The composition of the as-plated Fe-Pt film was measured with a scanning electron microscope-energy dispersive X-ray spectroscopy (SEM-EDX) system (Hitachi High-technologies S-3000). We measured nine points in the films, and the averaged value was shown as the film's composition. The hysteresis loops of the two-step-annealed Fe-Pt films were obtained using a vibrating sample magnetometer (VSM). The maximum applied field for the loop measurements was approximately 2 MA/m (25 kOe), and the remanence and coercivity values were obtained from the measured loop. To discuss the structural change of the Cu sheet (Cu substrate) by the LT-A, we obtained the X-ray diffraction (XRD) patterns of the Cu sheet using an X-ray diffractometer with $\text{Cu-K}\alpha$ radiation (Rigaku, Miniflex600-DX).

3. Results and Discussion

Figure 2 shows the remanence and coercivity of the two-step-annealed Fe-Pt films as a function of the holding time of LT-A. As shown in Fig. 2, the coercivity was almost the same value of approximately 900 kA/m, indicating that LT-A does not affect the coercivity. The remanence increased by increasing the holding time to ten minutes, then decreased. When considering the application

of Fe-Pt films as small magnets, high remanence is preferable. Our result suggests that moderate LT-A is effective in obtaining high remanence.

Rolling is a widely used metal-forming process, and Cu sheets are generally produced by rolling methods. As the rolling process causes permanent deformation of the Cu sheets, a large amount of rolling strain was expected to remain in them. We considered that LT-A releases the strain, causing a structural change that affects the remanence value. We, therefore, decided to obtain XRD patterns of the Cu substrate before and after LT-A to confirm the structural change.

Figure 3 shows XRD patterns of the Cu sheet before and after LT-A. We applied LT-A (300°C, 10 min) to the Cu sheet (without HT-A in Fig. 3). As shown in Fig. 3, although the diffraction peak from the (220) plane was strong before LT-A, the peak for the (111) plane became stronger after LT-A. This result implies that LT-A changes

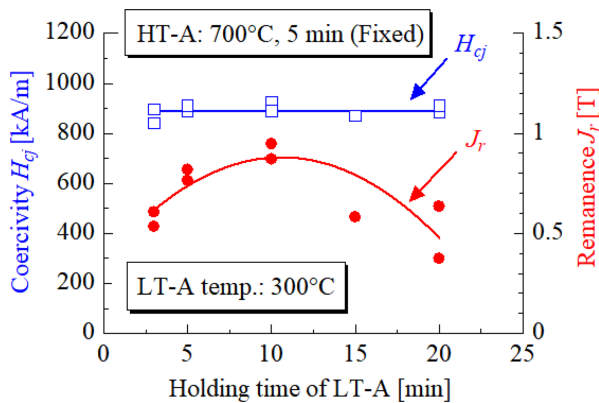


Fig. 2. (Color online) Remanence and coercivity of the two-step-annealed Fe-Pt films as a function of the holding time of LT-A.

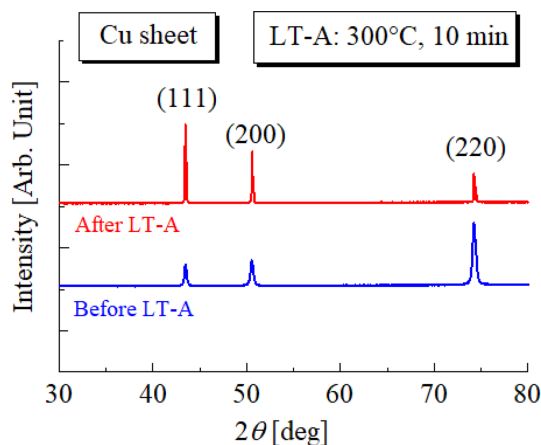


Fig. 3. (Color online) XRD patterns of the Cu sheet before and after LT-A.

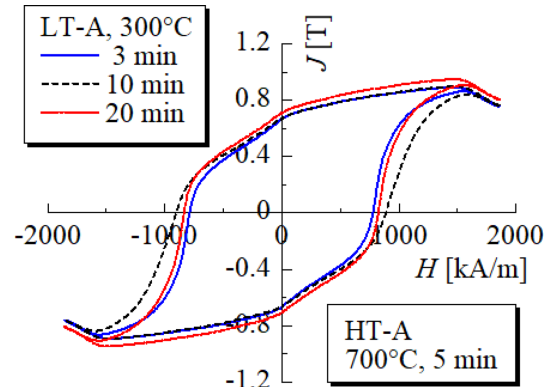


Fig. 4. (Color online) Hysteresis loops of the two-step-annealed Fe₅₀Pt₅₀ films electroplated on the 300°C-annealed Cu substrates.

the crystalline structure of the Cu substrate.

To confirm that the remanence variation by the LT-A is attributed to the structural change of the Cu substrate, we electroplated Fe-Pt films on the 300°C-annealed Cu sheets and then applied the two-step-annealing.

Figure 4 shows hysteresis loops of the two-step-annealed Fe₅₀Pt₅₀ films electroplated on the 300°C-annealed Cu substrates. When using the annealed Cu substrates, no noticeable variation in remanence was observed. This result strongly suggests that the remanence variation is attributable to the structural change of the Cu substrate.

Remanence is an important index of hard magnetic materials. The present study implies that remanence can be controlled by using the structural changes of the Cu substrate, and we found an interesting method to control the remanence of the electroplated Fe-Pt films.

4. Conclusion

We investigated the effect of two-step-annealing on the hard magnetic properties of the Fe-Pt films electroplated on the Cu sheets. The results are summarized as follows.

(1) The remanence of two-step-annealed Fe-Pt films varies with the holding time of LT-A, and a high remanence value was obtained at LT-A conditions of 300°C for 10 min.

(2) 300°C-annealing changed the structure of the fcc Cu crystalline phase in the substrate. Our results suggest that the remanence variation by LT-A is attributed to this structural change of the Cu substrate.

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