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Investigation of coercivity for electroplated Fe-Ni thick films

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We have already reported Fe-Ni films with good soft magnetic properties prepared by using an electroplating method. In our previous studies, we prepared the Fe-Ni films from citric-acid-based baths (CA-baths) and ammonium-chloride-based ones (AC-baths), and confirmed that the coercivity for the AC-baths was lower than that for the CA-baths. In the present study, we investigated reasons for the lower coercivity for the AC-baths to further improve the soft magnetic properties. From an observation of magnetic domains of the Fe₂₂Ni₇₈ films, we found that Fe₂₂Ni₇₈ film for AC-bath had a magnetic anisotropy in the width direction, and also found that the coercivity in the width direction was lower than the longitudinal one for the AC-bath. As an annealing for a stress relaxation in the films reduced the difference in the coercivity, we considered that the anisotropy is attributed to the magneto-elastic effect. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.5007293>

I. INTRODUCTION

Fe-Ni thick films with good soft magnetic properties have applied in electric devices such as magnetic sensors.¹⁻⁴ For fabrication of the thick films, electroplating methods are one of attractive techniques due to the high economic viability of the process. We reported Fe-Ni films with good soft magnetic properties prepared by an electroplating method,⁵ as well as many researchers reported.⁶⁻¹⁰ In our previous studies, we employed two types of plating baths, i.e. citric-acid-based baths (CA-baths)⁵ and ammonium-chloride-based ones (AC-baths)¹¹ for obtaining the Fe-Ni films. For typical electroplating of Ni and Fe-Ni films, plating baths with boric acid are widely used. We firstly employed the citric acid as a complex agent instead of the boric acid since waste baths containing boron are restricted by the regulation of environmental protection in Japan, and obtained Fe-Ni films with good soft magnetic properties. Although the baths with the citric acid enable us to obtain the films with good soft magnetic properties, current efficiency of the plating process was relatively low ($\approx 90\%$). Complex agents are often used for electroplating of metal films, and affect the current efficiency as well as various properties of the films, such as surface conditions, crystalline structures, and magnetic properties. Therefore, we secondly employed ammonium chloride as a complex agent to improve the magnetic properties and the current efficiency. Consequently, we found out that the AC-baths are also hopeful to obtain Fe-Ni films with good soft magnetic properties and high current efficiency of the process ($>95\%$), and confirmed that much lower coercivity of Fe₂₂Ni₇₈ films for the AC-baths compared with that for the CA-ones. In the present study, we investigated reasons for the lower coercivity for the AC-baths to further improve the soft magnetic properties.

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II. EXPERIMENTAL PROCEDURE

A. Electroplating of Fe-Ni films

We carried out an electroplating to obtain Fe-Ni films by using a direct current. The composition of the plating bath and the plating conditions are shown in Tables I and II, respectively.

B. Annealing

Since it is well-known that electroplated films have internal stress, we employed an annealing for stress relaxation of the as-plated Fe-Ni films. The films were annealed at 300°C for 60 min in a vacuum ($<10^{-2}$ Pa). The temperature was ramped from room temperature to the annealing temperature for 5 min, and then kept at constant for 60 min.

C. Measurements

The dc-hysteresis loops were measured with a B-H tracer (Riken Denshi BHS-40) operated at a field sweep rate of 50 mHz. The maximum excitation field of approximately 4 kA/m was used for the B-H measurements, and the coercivity values were obtained for the measured loops. The compositions of the films were analyzed by SEM-EDX (scanning electron microscope-energy dispersive X-ray spectroscopy: Hitachi High-technologies S-3000). The thicknesses of the as-plated Fe-Ni films were measured with a micrometer (Mitutoyo, CPM15-25MJ). As we fixed the current density and the plating time, all films showed almost the same thickness (Approx. 20 μ m). The magnetic domains of the Fe₂₂Ni₇₈ films were observed using a Magneto-Optical Kerr Effect (MOKE) microscope.

TABLE I. Components in the plating bath.

(a) AC-baths	
Components	Concentration
NiSO ₄ ·6H ₂ O (Nickel sulfate)	275 g/L
FeSO ₄ ·7H ₂ O (Iron sulfate)	25-100 g/L
C ₇ H ₄ NNaO ₃ ·2H ₂ O (Saccharin)	10 g/L
NH ₄ Cl (Ammonium chloride)	15 g/L
NaCl (Sodium chloride)	0-100 g/L
NaC ₁₂ H ₂₅ SO ₄ (Sodium lauryl sulfate)	5 g/L
C ₆ H ₈ O ₆ (Ascorbic acid)	3 g/L
(b) CA-baths	
Components	Concentration
NiSO ₄ ·6H ₂ O (Nickel sulfate)	275 g/L
FeSO ₄ ·7H ₂ O (Iron sulfate)	20-100 g/L
C ₆ H ₈ O ₇ ·7H ₂ O (Citric acid)	10 g/L
C ₇ H ₄ NNaO ₃ ·2H ₂ O (Saccharin)	5 g/L
NaCl (Sodium chloride)	50 g/L

TABLE II. Plating conditions.

Conditions	Values
Bath temperature	60°C
pH (Not adjusted)	Approx. 3.4 (AC bath) Approx. 2 (CA bath)
Current density	200 mA/cm ²
Plating time	5 - 40 min
Anode (Ni)	15 × 5 mm ² , 15 × 15 mm ²
Substrate(Cu)	15 × 5 mm ² , 15 × 15 mm ²

III. RESULT AND DISCUSSION

As shown in INTRODUCTION, $\text{Fe}_{22}\text{Ni}_{78}$ films prepared from the AC-baths showed lower coercivity values compared with those for the CA-ones. To discuss the difference in the coercivity between the AC-baths and the CA-ones, we observed the magnetic domains of the $\text{Fe}_{22}\text{Ni}_{78}$ films. Figure 1 shows the domain imaging of the $\text{Fe}_{22}\text{Ni}_{78}$ films (15 mm in length \times 5 mm in width) in remanence state. We observed clear contrast in the film prepared from the AC-bath, indicating that a magnetic anisotropy is induced in the width direction.

To confirm the effect of the anisotropy on the coercivity, we electroplated square-shaped $\text{Fe}_{22}\text{Ni}_{78}$ films (15 mm \times 15 mm), and evaluated the coercivity in the width direction and in the longitudinal one. Figure 2(a) shows coercivity in width and longitudinal directions of the Fe-Ni films prepared from the AC-baths. Typical hysteresis loops are also shown in Fig. 2. We describe the coercivity in each direction as wH_c and LH_c in this paper. The result for the CA-baths is also shown as FIG. 2(b). As shown in FIG. 2(a), wH_c was lower than LH_c in wide Fe content range, and we considered that the difference in the coercivity between width direction and longitudinal one is attributed to an anisotropy. The difference in the coercivity for the CA-bath FIG. (2(b)) is small, and the result for FIG. 2, which is not clear contrast of the domain image, agreed with that for FIG. 1 qualitatively.

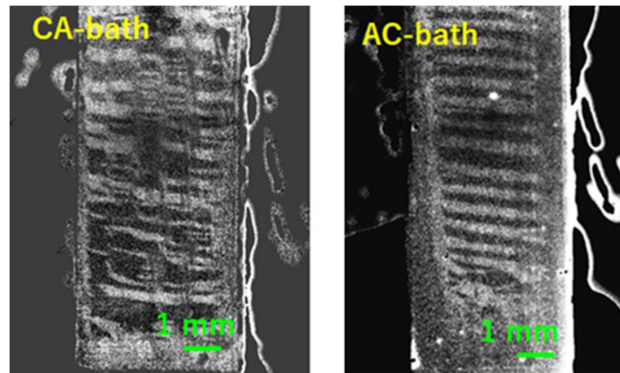


FIG. 1. Magnetic domain images of the $\text{Fe}_{22}\text{Ni}_{78}$ films (15 mm in length \times 5 mm in width) for the CA-bath and the AC-one.

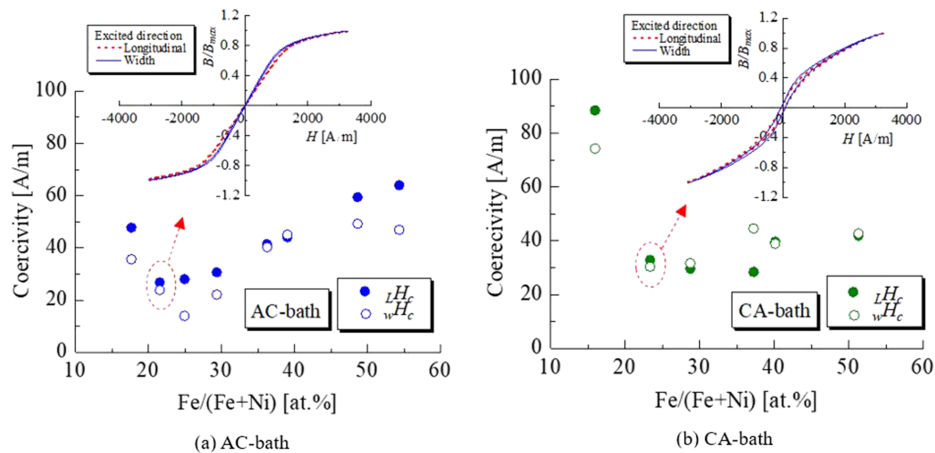


FIG. 2. Coercivity of the square-shaped Fe-Ni films (15 mm in length \times 15 mm in width) for (a) AC-bath and (b) CA-one as a function of the Fe content in the film. Typical hysteresis loops of the Fe-Ni films for the AC-bath and the CA-one are also shown in the figure.

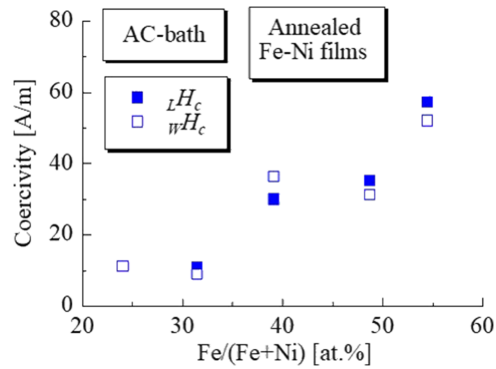


FIG. 3. Coercivity of the annealed Fe-Ni films as a function of Fe content.

Since typical electroplated films contain residual stresses, we considered that the observed anisotropy is attributed to a magneto-elastic effect. If the anisotropy was induced through the magneto-elastic effect in the as-plated Fe-Ni films, it is expected that a relaxation of the stresses reduces the difference in the coercivity. We, therefore, investigated an annealing for the stress relaxation.

Figure 3 shows coercivity of the Fe-Ni films annealed at 300°C as a function of the Fe content in the films. It is obvious that the difference in the coercivity is reduced by the stress-relaxation annealing, and we considered that the anisotropy shown in Fig. 1 is attributed to the magneto-elastic effect.

Since a magneto-elastic anisotropy energy in a magnetic material is proportional to magnitude of a stress, we expected that large residual stress in the as-plated films generates large difference in the coercivity. For electroplating of Ni films, it is well-known that chlorides in plating bath changes magnitude of an internal stress of the Ni films. For example, Turu *et al.* investigated the effect of chloride ions on the internal stress for electroplated Ni films, and reported difference in internal stress between plating baths with and without chloride ions is approximately 40 MPa for 5 μ m-thick Ni film.¹² Thus, we decided to vary the amount of sodium chloride (NaCl) in the AC-bath, and evaluate the difference in the coercivity. Figure 4 shows coercivity of the Fe₂₂Ni₇₈ films as a function of NaCl concentration. As shown in Fig. 4, the difference in the coercivity reduced with increasing the NaCl concentration. As it is considered that the amount of chloride (NaCl) affects the magnitude of the internal stress in the as-plated film, we found that the induced anisotropy in the width direction is attributed to the magneto-elastic effect, and also found that low coercivity can be obtained by exciting in the width direction of the films prepared from the baths with low NaCl concentration.

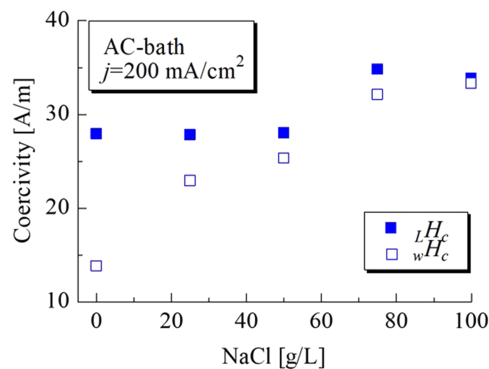


FIG. 4. Coercivity of the Fe₂₂Ni₇₈ films as a function of NaCl concentration.

IV. CONCLUSION

In conclusion, we have investigated the coercivity of the Fe-Ni films prepared from the AC-baths to further improve the soft magnetic properties. The obtained results are summarized as follows;

- (1) From the magnetic domain observation of the Fe₂₂Ni₇₈ films, a magnetic anisotropy, which is induced through magneto-elastic effect, was observed in the width direction for the AC-bath.
- (2) The coercivity in width direction was lower than that in longitudinal one in the wide Fe content range for the AC-bath due to the anisotropy.
- (3) The Chloride (NaCl) in the plating bath affects the magnitude of the difference in the coercivity between width direction and longitudinal one, and excitation in width direction of the film is effective to obtain low coercivity.

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- ¹ H.-S. Park, J.-S. Hwang, W.-Y. Choi, D.-S. Shima, K.-W. Na, and S.-O. Choi, *Sens. Act. A: Phys.* **114**, 224 (2004).
- ² T. M. Liakopoulos and C. H. Ahn, *Sens. Act. A: Phys.* **77**, 66 (1999).
- ³ P. Ripka, "Review of fluxgate sensors," *Sensors and Actuators A* **33**, 129 (1992).
- ⁴ Y. Watanabe, H. Nishizawa, H. Nakajima, M. Shikai, and T. Hirai, *IEEJ Trans. PE* **133**(11), 851 (2013).
- ⁵ T. Shimokawa, T. Yanai, K. Takahashi, M. Nakano, K. Suzuki, and H. Fukunaga, *IEEE Trans. Magn.* **48**, 2907 (2012).
- ⁶ M. Theis, S. Ediger, M. T. Schmitt, J.-E. Hoffmann, and M. Saumer, *Phys. Status Solidi A* **210**, 853 (2013).
- ⁷ Y. Kamo and T. Miyamoto, *IEEE Trans. Magn. Jpn.* **1**, 502 (1985).
- ⁸ D. Flynn and M. P. Y. Desmulliez, *IEEE Trans. Magn.* **46**, 979 (2010).
- ⁹ J. M. Quemper, S. Nicolas, J. P. Gilles, J. P. Grandchamp, A. Bosseboeuf, T. Bourouina, and E. Dufour-Gergam, *Sens. Act. A: Phys.* **74**, 1 (1999).
- ¹⁰ E. R. Spada, L. S. de Oliveira, A. S. da Rocha, A. A. Pasa, G. Zangari, and M. L. Sartorelli, *J. Magn. Magn. Mater.* **272**, e891 (2004).
- ¹¹ T. Yanai, K. Koda, K. Eguchi, K. Takashima, T. Morimura, M. Nakano, and H. Fukunaga, "Effect of ammonium chloride in plating baths on soft magnetic properties of electroplated Fe-Ni films," *IEEE Trans. Magn.* **53**, 2004303 (2017).
- ¹² Y. Tsuru, M. Nomura, and F. R. Foulkes, "Effects of chloride, bromide and iodide ions on internal stress in films deposited during high speed nickel electroplating from a nickel sulfamate bath," *J. Appl. Electrochem.* **30**, 231 (2000).