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Improvement in Current Efficiency of Gel-plating Process for Soft Magnetic Ni and Co Films

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Recently, we reported Ni films prepared from gel electrolytes and showed the potential of the gel-plating process. Our previous study confirmed that the saturation magnetization of the Ni films was smaller than that of bulk Ni, indicating that we need to improve the saturation magnetization for gel-plating. The present study focuses on improving the current efficiency of the gel-plating process and investigates the effects of the current density during the electroplating and the metal reagent's concentration in the baths on the efficiency and the saturation magnetization of Ni and Co films. From the results of the investigations for the current density, we confirmed that low current density improves the current efficiency of the process and increases the saturation magnetizations of Ni and Co films. Increasing the concentration of metal reagents also improves current efficiency and increases saturation magnetizations. From these results, we found that improvement in the diffusion limitation of metal ions, such as reduction in the current density and increase in the metal ion concentration, can enhance the current efficiency of the gel-plating process. The improvement in the efficiency increased the saturation magnetization, and we also found that an improvement in efficiency leads to an improvement in the magnetic properties.

Keywords : electroplating, soft magnetic films, gel electrolytes, magnetic properties, current efficiency, current density

1. Introduction

Commercial soft magnetic films are widely prepared using dry and wet processes. Wet processes, such as electroplating and electroless plating methods, are generally preferable over dry processes due to the simplicity of the equipment.

Fe-Ni and Fe-Co alloys are well-known as excellent soft magnetic materials, and their electroplated films have been widely studied [1-7]. We have also employed the electroplating method to obtain soft magnetic films and reported their good soft magnetic properties [8-11].

In recent years, environmental awareness has increased, and consideration of the environmental impact of fabrication processes is required. Waste reduction is necessary to reduce the environmental impact of electroplating processes. Therefore, we focused on electroplating processes using gel electrolytes (gel-plating) [12]. Our previous study employed a gel-plating method to obtain soft magnetic Ni films and found that soft magnetic Ni films could be obtained from the gel electrolyte [13]. Although gel-plating is one of the attractive fabrication processes of the Ni films, the saturation magnetization of the Ni films was smaller than that of bulk Ni [13], indicating that we need to improve the soft magnetic properties. The present study, therefore, focused on the improvement in the current efficiency of the plating process, which is the ratio of the electroplated film's weight to the theoretical one obtained by Faraday's law, and investigated the effects of the current density during the electroplating and the metal reagent's concentration in the baths on the efficiency and the saturation magnetization of Ni and Co films.

2. Experimental Procedures

2.1. Preparation of gel electrolyte

We electroplated Ni and Co films (single magnetic phase films) using gel-plating methods to remove the effect of the film's composition on magnetic and

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Table 1. Bath conditions.

Reagents	Concentration (g/L)
$NiSO_4 \cdot 6H_2O$ or $CoSO_4 \cdot 7H_2O$	60-220
Glycine	15
NaCl	2
Sodium naphthalene tri-sulfonate	2
pH	3-5 (Not adjusted)

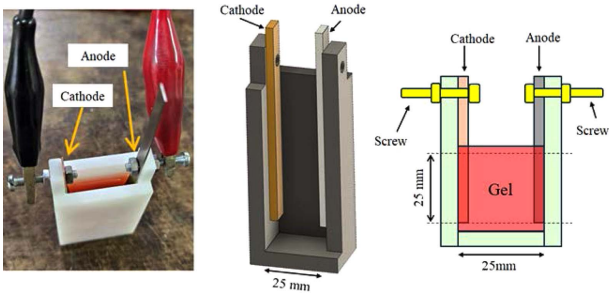

Fig. 1. (Color online) Jig for electroplating using gel electrolyte.

structural properties. Table 1 shows the bath conditions. First, we measured each reagent's weight based on the target concentrations in Table 1 and placed the reagents in a beaker. We added distilled water into the beaker to adjust the total mass to 50 g. The solution (distilled water with the reagents) was stirred at 50 °C for enough time, and gelatin (2 % of the total mass) was added. The solution with gelatin was stirred at 50 °C again. After completely dissolving the gelatin, we added the solution to the plating jig (Fig. 1) and cooled the jig with the solution in a refrigerator for 120 minutes to obtain a gel-electrolyte.

2.2. Electroplating from gel electrolyte

We electroplated Ni and Co films using a DC power supply. Fig. 1 shows a jig's photograph and schematic representations of the jig's inside. We placed the Fe plate (25 mm × 5 mm × 0.5 mm) as an anode and the Cu one (25 mm × 5 mm × 0.5 mm) as a substrate with a gap of 25 mm. The space between the electrodes was filled with gel electrolytes. Table 2 shows plating conditions. The bath temperature before the plating was approximately 15 °C, and the gel electrolyte was not heated during the plating to avoid the thermal decomposition of the gel electrolyte.

2.3. Measurements

The hysteresis loops were measured with a vibrating sample magnetometer, and the maximum applied field was approximately 1.5 MA/m. The current efficiency (C.E.) was calculated from the actual weight (W_{actual}) of the electroplated film and the theoretical weight (W_{theory}) obtained by Faraday's law. The related equations are as follows.

Table 2. Gel plating conditions.

Conditions	Values
Plating time t	10 min
Plating area S	125 mm^2 (25 mm × 5 mm)
Current density j	10-50 mA/cm^2

$$W_{theory} = \frac{MIt}{zF} \tag{1}$$

$$\text{C. E.} = \frac{W_{actual}}{W_{theory}} \tag{2}$$

Where M is the atomic weight, Z is the ion valence, I is the current for electroplating ($I = j \times S$), t is the plating time, and F is the Faraday constant (96485 C/mol).

For example, in the electroplating process of Ni films, the following reactions occurred on the cathode surface.

$$Ni^{2+} + 2e^- = Ni \tag{3}$$

$$2H_2O + 2e^- = H_2 + OH^- \tag{4}$$

As we need soft magnetic Ni (or Co) films, the reaction of Eq. (4) is not preferable. The current efficiency indicates the ratio of Eq. (3) in the plating process, and a high value is generally required.

3. Results and Discussion

3.1. Current density

Current density is an important parameter for electroplating, determining the film's quality and deposition rate. Therefore, we decided to investigate the effects of current density on current efficiency as a first step.

Figure 2 shows the current efficiency of the plating processes for Ni and Co films as a function of the current density. In this experiment, the metal reagents' concentrations were fixed at 60 g/L. As shown in Fig. 2, the current efficiency decreased with increasing current density. This result indicates that Eq. (4) becomes dominant when applying high current density. As shown in Eq. (4), the water decomposition generates H_2 gas and OH^-. When a large amount of OH^- is generated, hydroxides or oxides of Ni and Co are deposited in the films, which is expected to deteriorate the soft magnetic properties. To confirm the magnetic properties of the as-plated Ni and Co films, we evaluated the films' hysteresis loops. The obtained loops are shown in Fig. 3. For both

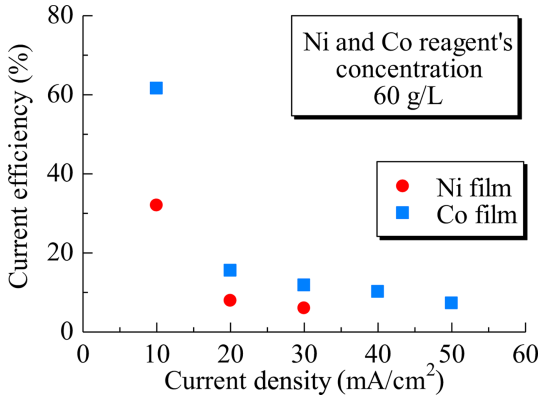

Fig. 2. (Color online) Current efficiency of the gel-plating processes for Ni and Co films as a function of current density during plating.

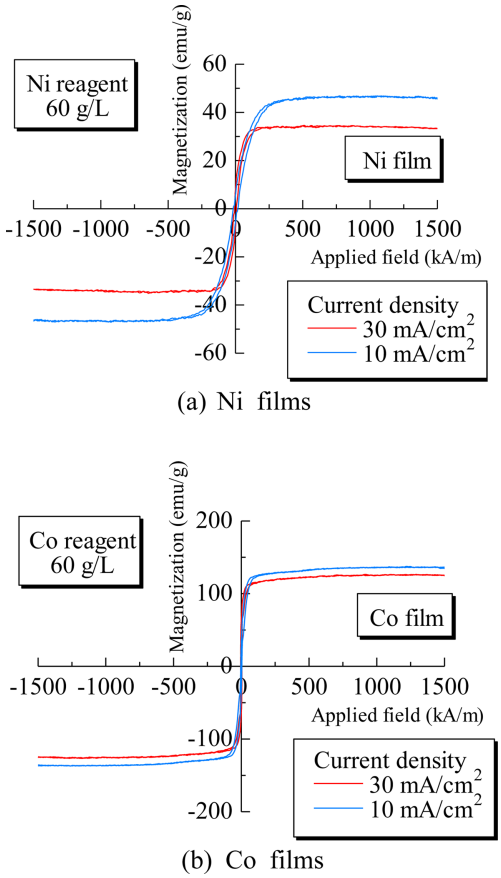

Fig. 3. (Color online) Hysteresis loops of the as-plated Ni and Co films electroplated at j = 10 and 30 mA/cm^2.

films, the saturation magnetization values for j = 30 mA/cm^2 are lower than those for j = 10 mA/cm^2, indicating that low current density effectively improves the soft magnetic properties. Although further reducing current density could enhance current efficiency and improve

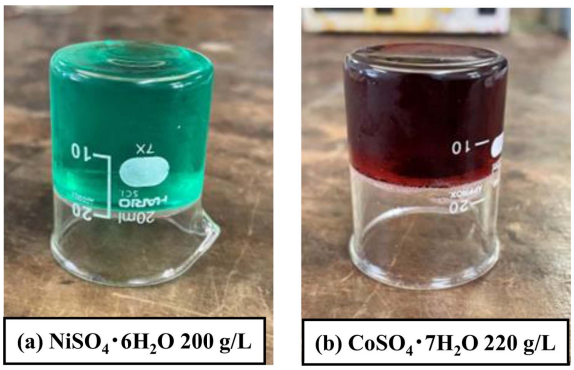

Fig. 4. (Color online) Photographs of the cooled electrolyte for (a) Ni (200 g/L) and (b) Co (220 g/L) plating.

saturation magnetization, low current density is undesirable for practical applications due to the dramatic decrease in deposition rate. Therefore, in Section 3.2, we investigated the effects of increasing the metal reagent's concentration in the plating bath.

3.2. Metal reagent's concentration

From the result in Fig. 2, we considered that the diffusion speed of metal ions in the gel electrolyte is not fast. Due to low diffusion speed, the lack of metal ions on the cathode surface rapidly increases at high current density, resulting in the reaction of Eq. (4) becoming dominant. To further supply the metal ions on the cathode (to reduce the lack of metal ions), we, therefore, investigated increasing the metal reagent's concentration in the plating baths and evaluated the effect of the concentration on the current efficiency.

As a preliminary study, we investigated the maximum concentrations of the Ni and Co reagents to obtain fully solidified electrolytes. Fig. 4 shows the photographs of the cooled baths in a refrigerator for 120 minutes. The green and red parts inside the breakers are the gel electrolytes for Ni and Co film's plating, respectively. Although the beakers were placed upside down in Fig. 4, the electrolytes kept their shape without falling. In our experimental conditions, the maximum concentrations were 200 g/L for the Ni plating and 220 g/L for the Co one, and a much higher concentration could not solidify the baths.

Figure 5 shows the current efficiency as a function of the Ni and Co reagent's concentration. Based on the investigations in Fig. 4, the concentrations were varied up to 200 g/L for Ni films and 220 g/L for Co ones, and the results for (a) j = 10 mA/cm^2 and (b) j = 20 mA/cm^2 are shown in Fig. 5.

As shown in Fig. 5, although the current efficiencies for

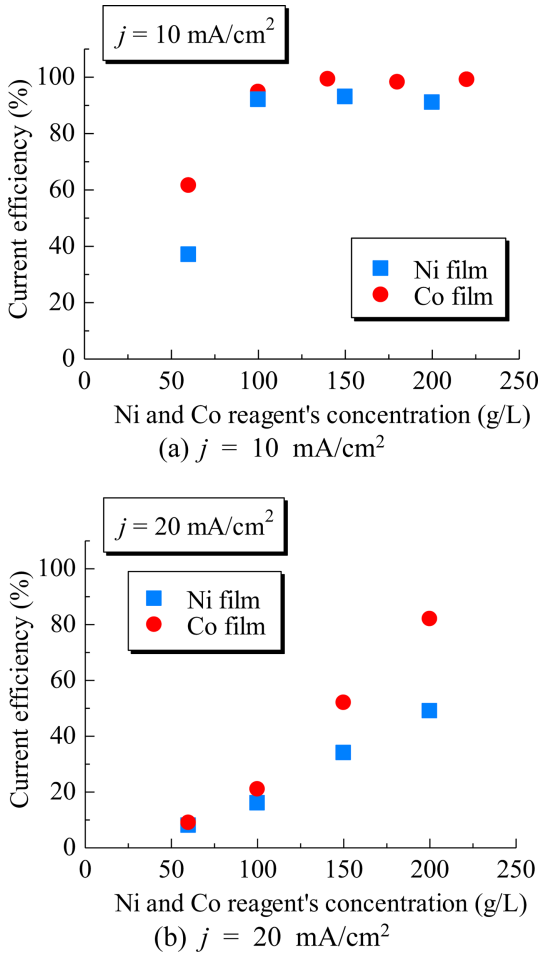

Fig. 5. (Color online) Current efficiency as a function of Ni and Co reagent's concentration at (a) j = 10 mA/cm^2 and (b) j = 20 mA/cm^2.

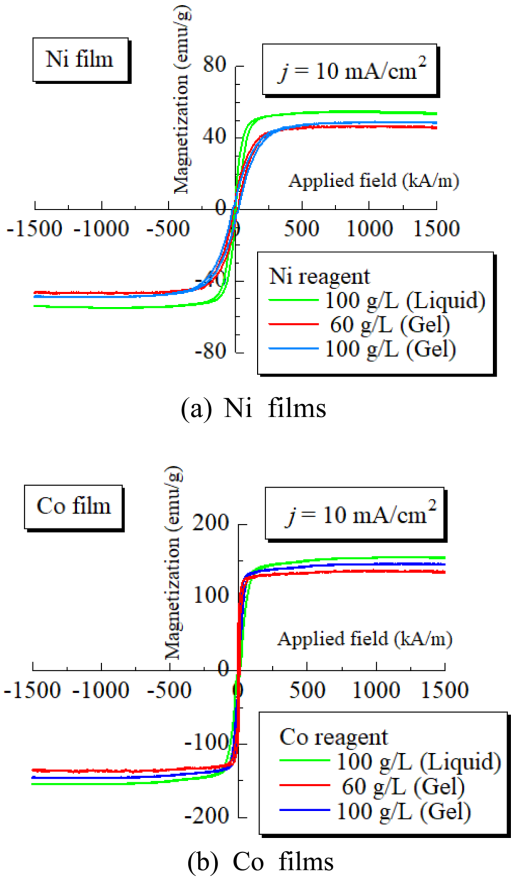

Fig. 6. (Color online) Hysteresis loops of (a) Ni and (b) Co films fabricated in the gel-based baths with the concentrations of 60 and 100 g/L. The loops for the liquid-based ones are also shown in this figure.

both Ni and Co films showed low values at j = 20 mA/cm^2, high current efficiencies (> 90 %) were obtained at j = 10 mA/cm^2 in the high concentration region (> 100 g/L). This result suggests that increasing the metal reagent's concentration is useful to improve current efficiency under a low current density. Considering the plating process's deposition rate and current efficiency, we found that 10 mA/cm^2 is a suitable current density for gel-plating in our experimental conditions.

Figure 6 shows the hysteresis loops of Ni and Co films fabricated in the gel-based baths with the concentrations of 60 and 100 g/L. For comparison, the loops for the liquid-based baths are also shown in Fig. 6. As mentioned in Fig. 5, we confirmed that high current efficiency could be obtained at high concentrations (> 100 g/L). From the viewpoint of saving the reagents, the results at 100 g/L are shown in Fig. 6.

As shown in Fig. 6, the saturation magnetization values of Ni and Co films slightly increased with increasing concentrations, and we found that a high metal reagent concentration effectively enhances the soft magnetic properties. However, the saturation magnetizations for the gel-based baths were smaller than those for the liquid-based ones. The decrease in saturation magnetization suggests that the films contain many impurities and oxidation of the magnetic element, and improving the saturation magnetization is one of our future works.

The results of the present study show that low current density and a high metal reagent concentration are suitable for preparing soft magnetic films using the gel plating method. This suggests that improving the diffusion limitation of metal ions, such as reducing the current density and increasing the metal ion concentration, can enhance the current efficiency of the gel-plating process.

4. Conclusion

The present study investigated the improvement in the magnetic properties of the electroplated Ni and Co films using the gel-plating method, focusing on the current efficiency. The results obtained are summarized as follows:

(1) The current efficiency was improved by reducing the current density and increasing the metal reagent's concentration in the plating baths.

(2) High current efficiency (90–95 %) was obtained at a high Ni and Co reagent concentration (100–200 g/L) at j = 10 mA/cm^2.

(3) Improvement in current efficiency enhances the saturation magnetization of the films.

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