Influence Mechanism of (NH$_4$)$_2$SO$_4$ on the Composition and Structure of Fe-Co Alloys

Yan Wu, Xue Qiao and Wei Wang *

School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, China
* Correspondence: wangweipaper@tju.edu.cn

Abstract: Fe-Co alloys have the advantages of high saturation magnetization and high magnetic elongation. They are used in electromagnet pole heads, earphone vibration films, magnetostrictive transducers, and so on. At present, few papers have been published on the co-deposition process of Fe-Co alloys. In the present work, smooth dense Fe-Co films of 10 µm were successfully prepared in an economical, green, sulfate solution system via electrodeposition. The cathodic polarization curves were measured separately under different conditions, and the electrochemical analyses showed that the increase in the main salt concentration is beneficial for obtaining higher deposition rates. An increase in temperature can increase the current density of electrochemical reduction of Fe$^{2+}$, and 50 °C is a suitable temperature at which to prepare the Fe-deposited layer with a flat surface. The increase in Co$^{2+}$ concentration can favor the co-electrodeposition process of Fe-Co alloys. Secondly, the EDS results showed that the mass fraction of Co in the films increased linearly with the increase in Co$^{2+}$ concentration in the solution. The mass fraction of Co in the films gradually increased with the increase in current density. The mass fraction of Co in the films gradually decreased with the increase in (NH$_4$)$_2$SO$_4$ concentration. In addition, the XRD showed that the grain size of the Fe-Co alloy films was closely related to the concentration of (NH$_4$)$_2$SO$_4$ in the electrolyte, and the smallest crystallite size of the Fe-Co alloy film was 17 nm. SEM showed that (NH$_4$)$_2$SO$_4$ can significantly affect the surface morphology of the films, in which the cracks are obviously removed.

Keywords: iron-cobalt; electrodeposition; stress; mechanism

1. Introduction

Magnetostrictive materials are functional for energy and information conversion. It has important applications in robots, sensors, displacement controllers, and other micro devices. Particularly, magnetostrictive materials can be found in microwave-absorbing films, smart electromagnetic films, magnetic recording films, magnetic core films for magnetic heads, etc. [1–4]. Magnetostrictive materials are divided into three main categories: materials containing rare earths, metals and alloys, and ferrite magnetostrictive materials [5,6]. Fe-Co alloys are known for their excellent magnetic flux density and low cost [7–9]. Nowadays, the production methods of Fe-Co alloys include both chemical and physical methods. Among them, chemical methods include electrodeposition, chemical deposition, etc., and physical methods include the vacuum vapor deposition method, the special rolling method, the vapor phase deposition technique, etc. [10–20]. Electrodeposition is an easy-to-operate, green, and economical technique for thin film fabrication. The advantages of electrodeposition are mild conditions; low time consumption; and the ability to control the size, thickness, and shape of the film by simply tuning the electrodeposition parameters.

Many efforts have been made by researchers to further improve the magnetic properties of Fe-Co alloys. The Fe-Co magnetic films prepared by E. A. Denisova et al. have excellent magnetic properties. Here, the saturation magnetic induction strength is about 2.47 T and the coercivity is greater than 1600 A/m [21]. R. S. Iskhakov et al. prepared Fe-Co alloys by electrodeposition in the chloride–sulfuric acid solution system using...
C$_3$H$_3$NaO$_3$S as an additive. The results of the study show that the coercivity of Fe-Co alloys is 400.02 A/m and the saturation magnetic induction intensity reaches 1.91 T for a C$_3$H$_3$NaO$_3$S concentration of 0.6 g/L [22]. Dong Zhou et al. investigated the effect of pulsed current electrodeposition on the magnetic properties of Fe-Co alloy films, and the results showed that the grain size of the samples prepared by pulsed electrodeposition was smaller than that of the samples prepared by the constant pressure method, which was conducive to the reduction of their coercivity [23]. In addition, magnetic nanowire arrays are valued for their potential advantages. These nanowires are widely used in perpendicular magnetic media due to their special characteristics, such as high magnetization strength and excellent perpendicular coercivity [24–28].

As often reported, it is not easy to control the growth and morphology of Fe-Co films. The obtention of magnetic films with high magnetic flux density and low coercive force is a key challenge, as these properties are usually difficult to combine because the requirements for film composition and crystal structure are different. Therefore, the process of exploring the influencing factors of the Fe-Co co-deposition process, understanding the mechanism of additives, and improving the formulation of the deposited Fe-Co film seems to be a promising method for adjusting the film’s properties [29,30]. In addition, S.V. Trukhanov et al. have shown that a good control of the stoichiometry of the alloy is important to the high quality of the alloys. Deviation from stoichiometry and the appearance of oxygen anions cause changes in the cation’s charge state, which in turn can dramatically alter the electronic, seriously affecting the practical application of the obtained materials [31,32].

In this study, smooth and dense Fe-Co films of 10 µm were prepared directly by electrodeposition through careful control of the electrodeposition parameters and electrolyte composition. The results showed that ammonium (NH$_4$)$_2$SO$_4$ as an additive significantly affected the co-deposition process of Fe-Co alloys and affected the composition, microstructure, and surface morphology of the alloys, and further affected the magnetic properties of the materials. Microscopic understanding of the co-electrodeposition process of Fe-Co alloys is still unsatisfactory. Therefore, the influence mechanism of (NH$_4$)$_2$SO$_4$ on the Fe-Co co-deposition process was thoroughly analyzed, including the concentration of Fe$^{2+}$ and Co$^{2+}$, the contents of (NH$_4$)$_2$SO$_4$, the ratios of Co$^{2+}$/(Fe$^{2+}$ + Co$^{2+}$), the current density, and the grain size. A strategy of physical dispersion (magnetic stirring) and chemical dispersion (adding surfactant) was used to promote the co-electrodeposition of Fe$^{2+}$ and Co$^{2+}$. The results are beneficial for guiding the Fe-Co co-deposition process, and helping us to reduce the internal stress so that a high-performance Fe-Co magnetic film can be produced. The study of these theoretical mechanisms can help us to prepare Fe-Co films with better magnetostrictive properties in our next work.

2. Experiment
2.1. Preparation of Fe-Co Alloys

In this study, a titanium plate with dimensions of 10 cm $\times$ 8 cm $\times$ 0.1 cm was used as the cathode substrate, the working area dimensions were 6 cm $\times$ 4 cm, and the non-electrodeposited area of the titanium sheet was sealed with epoxy resin. The titanium sheet was soaked in an acid solution for 10 s for activation before electroplating, and then rinsed in distilled water. The electrodeposition system consisted of an electrolytic cell (beaker with a volume of 2 L), an anode (titanium mesh), a cathode (titanium sheet), and a DC power supply. Schematic diagram of the Fe-Co electrodeposition process can be found in Figure 1. The iron-cobalt alloy was electrodeposited in an electrolyte containing 0.12 M FeSO$_4$·7H$_2$O, 0.06 M CoSO$_4$·7H$_2$O and a 0.18 M (NH$_4$)$_2$SO$_4$ group. The pH of the solutions was adjusted with dilute sulfuric acid and ammonium hydroxide, respectively. The Fe-Co films were electrodeposited at a current density of 50 mA/cm$^2$ for 13 min. The solution was stirred at 400 rpm during the electrodeposition process.
Films were electrodeposited at a current density of 50 mA/cm² for 13 min. The solution was stirred at 400 rpm during the electrodeposition process.

Figure 1. Schematic diagram of the Fe-Co electrodeposition process.

2.2. Electrochemical Characterization

Electrochemical tests were performed using an electrochemical workstation (CHI 660B). A platinum grid with an area of 12 cm² was used as the auxiliary electrode, a saturated glyceryl electrode (SCE) was used as the reference electrode, and a titanium sheet with an area of 1 cm² was used as the working electrode. A saturated calomel electrode was used relative to the given potentials and the scan rate of cathodic polarizing curves was 0.5 mV/s. X-ray diffraction (XRD) patterns were tested to determine the alloy structure and particle size. Finally, the morphology of the films was measured by SEM, and the composition and content of the film were analyzed by Energy Dispersive Spectrometer (EDS). The thickness was measured by the thickness meter with high precision (0.1 µm).

3. Results and Discussion

3.1. Electrodeposition Process of Fe-Co films

3.1.1. Analyses by Cathodic Polarization of Fe²⁺

The cathodic polarization curves of different Fe²⁺ concentrations at 50 °C were measured and shown in Figure 2a. From the figure, it can be seen that the current density gradually increases with the negative shift of the potential. According to Figure 2a, the relations between the current density and the concentration of Fe²⁺ when the potential is −1.2 V are shown in Figure 2b. It can be observed that the current density increases linearly with the increase in Fe²⁺ concentration. Additionally, the ratio of Fe²⁺ concentration to the cathodic current density |Ic| is 0.002, and the ratio is 0.001 as the Fe²⁺ concentration increases. The results indicate that the increase in Fe²⁺ concentration favors a higher deposition rate.

The cathodic polarization curves of 0.06 M Fe²⁺ at different temperatures are displayed in Figure 3a. From the figure, it can be observed that the current density gradually increases with the negative shift of the potential. The relations between the current density and the temperatures when the potential is −1.2 V in Figure 3a are shown in Figure 3b. The current density increases at an accelerated rate with the increase in the temperature. Additionally, the ratio of temperature to the cathodic current density |Ic| is 3.3, and the ratio is 1.1 as the temperature increases. The results indicate that the temperature affects the depositing rate of Fe²⁺ remarkably.
The cathodic polarization curves measured in solutions of 0.12 M Fe$^{2+}$ and different concentrations of (NH$_4$)$_2$SO$_4$ are shown in Figure 4a. It can be observed that the current density decreases for 0.05 M (NH$_4$)$_2$SO$_4$ at the same potential compared to the solution without (NH$_4$)$_2$SO$_4$, and after 0.05 M, the current density increases as the (NH$_4$)$_2$SO$_4$ concentration increases. The relationship between the (NH$_4$)$_2$SO$_4$ concentration and the current density at a potential of −1.2 V in Figure 4a is shown in Figure 4b. It can be observed that the addition of (NH$_4$)$_2$SO$_4$ in a low concentration, such as 0.05 M, reduces the current density. As the concentration of (NH$_4$)$_2$SO$_4$ is higher than 0.1 M, the current density increases with the increase in (NH$_4$)$_2$SO$_4$ concentration. The results exhibit that the high concentration of (NH$_4$)$_2$SO$_4$ accelerates the deposition while the addition of (NH$_4$)$_2$SO$_4$ in very small amounts inhibits the deposition. We believe that a small amount of (NH$_4$)$_2$SO$_4$ adsors at the electrode-solution interface and hinders the hydrogen evolution reaction in the initial stage, so the current density decreases. However, a large amount of (NH$_4$)$_2$SO$_4$ benefits the deposition of metal ions, so the current density increases [33].
The initial stage, so the current density decreases. However, a large amount of (NH₄)₂SO₄ sorbs at the electrode-solution interface and hinders the hydrogen evolution reaction in sorbs at the electrode-solution interface and hinders the hydrogen evolution reaction in increases. The results indicate that the increase in Co²⁺ concentration favors a higher deposition rate.

3.1.2. Analyses by Cathodic Polarization of Co²⁺

The cathodic polarization curves measured in solutions with different Co²⁺ concentrations are shown in Figure 5a. It can be observed that the current density gradually increases as the potential moves negatively. According to Figure 5a, the relation between the current density and the concentration of Co²⁺ when the potential is −1.2 V is shown in Figure 5b. From the figure, it can be observed that the current density increases linearly with the increase in Co²⁺ concentration. Additionally, the ratio of Co²⁺ concentration to the cathodic current density |Ic| is 0.001, and the ratio is 0.003 as the Co²⁺ concentration increases. The results indicate that the increase in Co²⁺ concentration favors a higher deposition rate.

The cathodic polarization curves tested with different (NH₄)₂SO₄ concentrations in the solution with 0.06 M Co²⁺ are shown in Figure 6a. The corresponding current density at a potential of −1.2 V is shown in Figure 6b. It can be observed from the figure that the current density decreases and then increases with the increase in the (NH₄)₂SO₄ concentration and reaches its minimum at the (NH₄)₂SO₄ concentration of 0.05 M. The current density of
(NH₄)₂SO₄ increases with the increase in (NH₄)₂SO₄ concentration. The results indicate that the electrolyte addition of an appropriate amount of (NH₄)₂SO₄ is beneficial for obtaining a higher Co²⁺ deposition rate.

3.1.3. Analyses by Cathodic Polarization of Fe²⁺ and Co²⁺

The cathodic polarization curves of different Co²⁺ concentrations are shown in Figure 7a. As can be seen from the figure, the current density gradually increases with the negative shift of the potential. From Figure 7b, it can be observed that the relationship between the proportion of Co²⁺ in the solution and the current density at a potential of −1.2 V. The results show that the current density gradually increases with the increase in the ratio of Co²⁺/(Fe²⁺ + Co²⁺). The growth rate of current density is greater when the ratio of Co²⁺/(Fe²⁺ + Co²⁺) is greater than 0.5 than that when the ratio of Co²⁺/(Fe²⁺ + Co²⁺) is less than 0.5. The above results indicate that increasing the proportion of Co²⁺ in the electrolyte can accelerate the electrochemical reaction rate of Fe²⁺ and Co²⁺, which is beneficial to the co-electrodeposition process. According to the Nernst Equation (1) [34], we calculated

$$E = E^0 - \frac{RT}{nF} \ln a$$  \hspace{1cm} (1)$$

where $E$ is the electrode potential at a certain concentration, $E^0$ is the standard electrode potential, $R$ is the gas constant (8.314 J·K⁻¹·mol⁻¹), $T$ is the temperature (K), $n$ is the net number of electrons gained and lost in the electrode reaction, $F$ is Faraday’s constant (96,485 C mol⁻¹).

Figure 8a shows the effect of (NH₄)₂SO₄ on the co-electrodeposition process of the Fe-Co alloy. The cathodic polarization curves in solutions with different (NH₄)₂SO₄ concentrations undergo a similar process of change. The current density gradually increases with the negative shift of the potential, and decreases when 0.05 M (NH₄)₂SO₄ is added to the electrolyte compared with the electrolyte without (NH₄)₂SO₄. Beyond this concentration, the current density increases monotonically with the (NH₄)₂SO₄ concentration [33]. The influence of the (NH₄)₂SO₄ concentration on Fe²⁺, Co²⁺, and the co-electrodeposition process are shown in Figure 4b, Figure 6b, and Figure 8b, respectively. From Figure 8b, it can be seen that the current density of Fe-Co alloy co-deposition is greater than that of Fe²⁺ and Co²⁺ electrodeposition alone. As the concentration of (NH₄)₂SO₄ in the electrolyte increases, the electrochemical reaction rates of the Fe-Co alloy, Fe²⁺, and Co²⁺ all decrease.
and then increase. When the concentration of \((\text{NH}_4)_2\text{SO}_4\) in the electrolyte was less than 0.13 M, the electrochemical reaction rate of \(\text{Fe}^{2+}\) was larger than that of \(\text{Co}^{2+}\), and the electrochemical reaction growth rate of \(\text{Co}^{2+}\) was faster than that of \(\text{Fe}^{2+}\), indicating that \((\text{NH}_4)_2\text{SO}_4\) could be more favorable for promoting the electrodeposition process of \(\text{Co}^{2+}\).

\[
\eta = \eta_{\text{Fe}} + \eta_{\text{Co}} = \frac{W_{\text{Fe}} \cdot \Delta G}{T \cdot I \cdot C_{\text{Fe}}} + \frac{W_{\text{Co}} \cdot \Delta G}{T \cdot I \cdot C_{\text{Co}}}
\]

To further investigate the effect of \((\text{NH}_4)_2\text{SO}_4\) on Fe-Co alloys, Figure 9 shows the effect of \((\text{NH}_4)_2\text{SO}_4\) on the current rate of the Fe-Co alloy. According to Equation (2) [35], the current efficiency \(\eta\) of the Fe-Co alloy was calculated. The study showed that the current efficiency of the Fe-Co alloy gradually increases with the increase in the \((\text{NH}_4)_2\text{SO}_4\) concentration in the electrolyte. When the \((\text{NH}_4)_2\text{SO}_4\) concentration is in the range of 0.05–0.2 M, the current efficiency of Fe-Co alloy grows at a greater rate. Additionally, when the \((\text{NH}_4)_2\text{SO}_4\) concentration exceeds 0.2 M, the current efficiency of the Fe-Co alloy grows slowly. Too much \((\text{NH}_4)_2\text{SO}_4\) is detrimental to the hydrogen precipitation reaction and internal stress; therefore, the study concluded that the suitable \((\text{NH}_4)_2\text{SO}_4\) concentration for the preparation of Fe-Co alloy is 0.18 mol/L.

**Figure 7.** (a) Cathodic polarization curves measured in solutions with different \(\text{Co}^{2+}\) concentrations; (b) the corresponding cathodic current density \(|I_c|\) at a potential of \(-1.2\) V.

**Figure 8.** (a) Cathodic polarization curves measured in solutions with different \((\text{NH}_4)_2\text{SO}_4\) concentrations; (b) the corresponding cathodic current density \(|I_c|\) at a potential of \(-1.2\) V.

**Figure 9.** The effect of \((\text{NH}_4)_2\text{SO}_4\) on Fe-Co alloys.
\( \eta_{Fe} \) and \( \eta_{Co} \) are the current efficiency of electrodeposited Fe and Co, respectively. \( C_{Fe} \) and \( C_{Co} \) are the electrochemical equivalents of Fe and Co, respectively. \( \Delta G \) is the mass of the alloy coating, \( W_{Fe} \) and \( W_{Co} \) are the mass percentages of Fe and Co in the alloy, respectively, \( I \) is the cathode deposition current, and \( t \) is the deposition time.

\[ \eta_{Co} = \frac{i_{Co^2+}}{i_{Co^2+} + i_{Fe^2+}} \]

**Figure 9.** Effect of different concentrations of (NH\(_4\))\(_2\)SO\(_4\) on the current efficiency of Fe-Co alloy.

### 3.2. Analyses of the Composition

The relation between \( i_{Co^2+}/(Co^{2+} + Fe^{2+}) \) and the mass fraction of Co in the film without (NH\(_4\))\(_2\)SO\(_4\) is shown in Figure 10a. The study shows that when the potential shifts negatively, the current density in the Fe\(^{2+}\) solution increases more than that in the Co\(^{2+}\) solution, so \( i_{Co^2+}/(Co^{2+} + Fe^{2+}) \) decreases gradually. The ion ratio used in \( i_{Co^2+}/(Fe^{2+} + Co^{2+}) \) is 33.3%. It can be seen that \( i_{Co^2+}/(Co^{2+} + Fe^{2+}) \) is significantly lower than the Co wt.% in the film (more than 35 wt.%). The results show that Fe\(^{2+}\) promotes the deposition of Co\(^{2+}\) in the solution without (NH\(_4\))\(_2\)SO\(_4\). The relation between \( i_{Co^2+}/(Co^{2+} + Fe^{2+}) \) and the mass fraction of Co in the film with (NH\(_4\))\(_2\)SO\(_4\) is shown in Figure 10b. The study shows that when the potential shifts negatively, the current density in the Co\(^{2+}\) solution is greater than that in the Fe\(^{2+}\) solution, and \( i_{Co^2+}/(Co^{2+} + Fe^{2+}) \) is significantly greater than the Co wt.% in the film. The results show that (NH\(_4\))\(_2\)SO\(_4\) inhibits the deposition of Co\(^{2+}\).

**Figure 10.** (a) Relations between \( i_{Co^2+}/(Co^{2+} + Fe^{2+}) \) and the mass fraction of Co in the film without (NH\(_4\))\(_2\)SO\(_4\); (b) Relation between \( i_{Co^2+}/(Co^{2+} + Fe^{2+}) \) and the mass fraction of Co in the film with (NH\(_4\))\(_2\)SO\(_4\).

The effect of the ratio of Co\(^{2+}/(Fe^{2+} + Co^{2+}) \) in the solution on the Co mass fraction in the films is shown in Figure 11a. It can be found the Co wt.% of the film increases linearly...
with the increase in the ratio of Co$^{2+}$/\((\text{Fe}^{2+} + \text{Co}^{2+})\) of the solution. Additionally, the ratio of Co$^{2+}$ concentration to the Co wt.% in the film is 0.99. The result indicates that anomalous deposition behavior is not observed here, in contrast to that observed in Fe-Ni alloy film deposition. The effect of the current density on the Co mass fraction in the film is shown in Figure 11b. It can be observed that the mass fraction of Co in the film linearly increases with the increasing current density below 40 mA/cm$^2$, and the Co content in the film is significantly slower beyond 40 mA/cm$^2$. Additionally, the ratio of current density to the mass fraction of Co in the film is 8.3, and the ratio is 15 as the current density increases. According to this standard, the desired composition for the purpose of achieving better properties can be controlled by the current density. The effect of \((\text{NH}_4)_2\text{SO}_4\) on the content of Co in the film is shown in Figure 11c. It can be observed that as the concentration of \((\text{NH}_4)_2\text{SO}_4\) increases, the content of Co in the film decreases gradually, and the content of Co in the film reaches its maximum at 0.5 M \((\text{NH}_4)_2\text{SO}_4\). Additionally, the ratio of the \((\text{NH}_4)_2\text{SO}_4\) concentration to the mass fraction of Co in the film is 1.67, and the ratio is $-1.19$ as the \((\text{NH}_4)_2\text{SO}_4\) concentration increases. The results illustrate that the concentration of \((\text{NH}_4)_2\text{SO}_4\) affects the composition of the film obviously, and may therefore affect the internal stress of Fe-Co films.

Figure 11. Variation curves of factors influencing the content of elemental Co in the films (a) Co$^{2+}$ concentration in the solution; (b) Current density; (c) \((\text{NH}_4)_2\text{SO}_4\) concentration.
3.3. Structure Analysis

Figure 12 shows the XRD patterns with and without the (NH$_4$)$_2$SO$_4$ in Fe$^{2+}$ solution, the Co$^{2+}$ solution, the Fe$^{2+}$ and Co$^{2+}$ solutions, which proves that the films prepared in this study are Fe-Co alloys. Electrodeposition is an epitaxial growth mode and ammonium sulphate directly affect the electrocrystallisation process following the electrochemical reduction of the metal ions, thus affecting the grain orientation and strength. As seen in Figure 12, (NH$_4$)$_2$SO$_4$ changes the orientation of the Co deposition, but does not change the orientation or strength of the Fe and FeCo alloys. Additionally, according to the half-peak width of the XRD patterns, the crystallite size was calculated by the Scherrer Formula (3), and the crystallite size with different (NH$_4$)$_2$SO$_4$ concentrations is shown in Figure 13. The study shows that as the concentration of (NH$_4$)$_2$SO$_4$ increases, the crystallite size of Fe increases, but the crystallite size of Co decreases. As for the film of Fe-Co alloy, the crystallite size decreases firstly and then increases, and the smallest crystallite size of Fe-Co alloy film is 17 nm when C$_{(NH4)2SO4} / C_{(Fe^{2+}+Co^{2+})}$ is 1.5. The results indicate that the crystallite size of Fe-Co alloy film can be controlled by the concentration of (NH$_4$)$_2$SO$_4$.

\[
D = \frac{(K\gamma)}{B\cos\theta}
\]  

(3)

where \(K\) is a constant, 0.943, for the cubic structure, \(\gamma\) is the X-ray wavelength, 0.15406 nm; \(B\) is the half-peak width of the diffraction peak, which is obtained by Jade; \(\theta\) is the diffraction angle.

Figure 12. XRD patterns with and without (NH$_4$)$_2$SO$_4$. (a) Fe$^{2+}$ solution; (b) Co$^{2+}$ solution; (c) Fe$^{2+}$ and Co$^{2+}$ solution.
Figure 13. Crystallite size with different (NH₄)₂SO₄ concentrations.

Finally, Figure 14 shows the morphology of the films with 10 µm thickness as measured by SEM. Additionally, the Co content in the film is 34 wt.%, and the oxygen content across the film thickness is about 2.45 wt.%. It is shown that a certain concentration of (NH₄)₂SO₄ can significantly affect the surface morphology of the film and eliminate cracks to obtain a dense and smooth film.

Figure 14. SEM photos of surface and section of Fe-Co films electrodeposited by (NH₄)₂SO₄. (a) Surface morphology without (NH₄)₂SO₄. (b) Surface morphology with (NH₄)₂SO₄. (c) Section morphology with (NH₄)₂SO₄.

4. Conclusions

It can be concluded that smooth dense Fe-Co films with 10 µm thickness were successfully prepared by the electrodeposition technique in a non-toxic sulfate solution system. Firstly, the cathodic polarization curves were measured with respect to different conditions at 50 °C, and pH 2.5. The results indicated that the increase in main salt concentration is beneficial to obtaining a higher depositing rate. Increasing the temperature can speed
up the electrochemical reaction of Fe\(^{2+}\); combining the effects of other aspects, it was determined that a temperature of 50 °C is suitable for the preparation of flat films. The increase in the proportion of Co\(^{2+}\) in the electrolyte favors the Fe-Co co-electrodeposition process. When the concentration of (NH\(_4\))\(_2\)SO\(_4\) in the electrolyte exceeded 0.13 M, the electrochemical reaction rate of Co\(^{2+}\) was greater than that of Fe\(^{2+}\), and (NH\(_4\))\(_2\)SO\(_4\) was more favorable to the electrochemical reaction process of Co\(^{2+}\). Secondly, the results of EDS showed that the Co content in the film increased linearly with the increase in the proportion of Co\(^{2+}\) in the solution. The Co content in the film gradually increased with the rise of the current density. As the concentration of (NH\(_4\))\(_2\)SO\(_4\) increased, the content of Co in the film decreased gradually. The XRD showed that the crystallite size of Fe-Co alloy films can be controlled by the concentration of (NH\(_4\))\(_2\)SO\(_4\), and the smallest crystallite size of Fe-Co alloy film was 17 nm. SEM showed that a certain concentration of (NH\(_4\))\(_2\)SO\(_4\) can eliminate the cracks on the surface of the films and result in smooth and dense flat films. These results provide evidence for the potential improvement of the Fe-Co co-deposition process and preparation of Fe-Co magnetic films with excellent performance in further research.

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**References**


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