# Electrodeposited CoNiFeP Soft-Magnetic Films for High-Frequency Applications

Fernando M. F. Rhen and Saibal Roy

Microsystems Center, Tyndall National Institute Lee Maltings, Prospect Row, Cork, Ireland

**We have studied the influence of P and of plating current density on static/dynamic magnetic and electrical properties of CoNiFe high moment alloy. We found that morphology, electrical and magnetic properties of films with P content up to 10 at. % are quite different from those of pure CoNiFe. CoNiFeP morphology consists of a structure made of isolated islands with sizes varying between 10 and 50 $\mu$m. We indirectly obtained resistivity in this structure by using high-frequency magnetic measurements. Film composition is weakly dependent on plating current density for values larger than 10 mA cm$^{-2}$. Coercivities increase up to 6400 A m$^{-1}$ and magnetization saturation is reduced to 1.0 T in CoNiFeP films. The most remarkable effect of P addition to CoNiFe is the increase of resistivity over two order of magnitude with values of 5.2 $\times$ 10$^{-5}$ $\Omega$ m compared to 2.4 $\times$ 10$^{-7}$ $\Omega$ m of pure CoNiFe. We also found that the ferromagnetic resonance of CoNiFeP alloy is 3 GHz for sample with 10 at. % P and a performance factor (BF) of $\sim$4 $\times$ 10$^5$ T s$^{-1}$, which is a better performance than bulk ferrite.**

*Index Terms*—**CoNiFeP, high resistivity, magnetic recording, power electronics.**

## I. Introduction

THE research in high moment alloy films has mostly been driven by applications in magnetic recording media, recording heads, microinductors, and microtransformers. The basic requirements for the materials for recording head and microinductor/microtransformer applications are high magnetization saturation, low coercivity, and high resistivity. High saturation magnetization is required to write on magnetic memory elements with high coercivity resulting in higher information density and to achieve high power density in case of microfabricated inductors/transformers. The most attractive alloy investigated to date is CoNiFe [1], which shows saturation magnetization as high as 2.1 T [2], [3]. However, due to low resistivity of CoNiFe (25 $\mu\Omega$ cm) the thickness is limited to submicron when higher operation frequency is required (>100 MHz).

Several approaches have been undertaken to increase resistivity, including the incorporation of sulphur [4], [5] and carbon [6] into the deposits. The available literature suggests that small elements such as S and C go to the grain boundaries and hence increase resistivity by enhancing electronic scattering between grains. However, as these elements have low atomic weight and are incorporated in such a small level (<1 at. %), it is very difficult to experimentally determine whether grain boundary segregation is present.

We have investigated the electrodeposition of CoNiFeP from a single electrochemical bath and studied the magnetic properties of this alloy. Phosphorous was incorporated to the bath aiming higher resistivity. We found that the incorporation of P increased resistivity and coercivity of CoNiFe films.

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## II. Experiment

CoNiFeP films were electrodeposited from an electrolyte as described in Table I. Test samples were electrodeposited from freshly prepared solutions, using strong mechanical agitation, in open atmosphere, at room temperature and using galvanic direct current (dc) current. Si substrates with a 200-nm sputtered Cu seed layer were varnished leaving an exposed Cu area of $5 \times 5$ mm$^2$ and used as substrates for electrodeposition of CoNiFeP. To remove any oxide layer on the substrate, they were washed in diluted sulphuric acid solution before deposition for a few seconds, and deposition was started immediately after immersion into the electrolyte. Microstructure, composition, thickness, and magnetic properties were determined using scanning electron microscope (SEM), energy dispersive spectroscopy (EDX), surface profilometer and a 5-T superconducting quantum interference device (SQUID) magnetometer, respectively. Films with different thicknesses were obtained by varying the deposition time. High-frequency permeabiltity spectra up to 9 GHz were determined using a Ryowa permeameter model PMM-9G1.

## III. Results and Discussion

The CoNiFeP electrolyte is very stable before and during the plating process. The electrodeposited CoNiFeP films are shiny and show small cracks. Films as thick as 30 $\mu$m were deposited. We have studied the dependence of CoNiFeP film composition on plating current density as shown in Fig. 1. The results show that the composition is weakly dependent on the current density for current densities larger than 10 mA cm$^{-2}$.

Room temperature magnetic properties of CoNiFeP were characterized using a SQUID magnetometer. Coercivity and magnetization saturation dependence on plating current density are shown in Fig. 2(a) and (b), respectively. In contrast to composition, the coercivity and saturation magnetization of CoNiFeP are strongly correlated to plating current density. The addition of P is known to result in higher coercivity in CoPt alloys. In this situation, P preferentially deposits around the grain boundaries and serves as pinning center, thereby

TABLE I
ELECTROLYTE COMPOSITION AND PLATING CONDITION FOR THE DEPOSITION OF CoNiFeP FILMS

| Chemical | Concentration mol $l^{-3}$ |
|---|---|
| $CoSO_4 - 7H_2O$ | 0.060 |
| $NiSO_4 - 7\,H_2O$ | 0.200 |
| $FeSO_4 - 7\,H_2O$ | 0.015 |
| $H_3BO_3$ | 0.400 |
| $NH_4Cl$ | 0.280 |
| $NaH_2PO_2$ | 0.025 |
| Sodium dodecyl sulfate | 10 mg $l^{-1}$ |
| Bath pH** | 2.8 |
| Plating condition | DC |
| Counter electrode | Co |

** adjusted with $H_2SO_4$

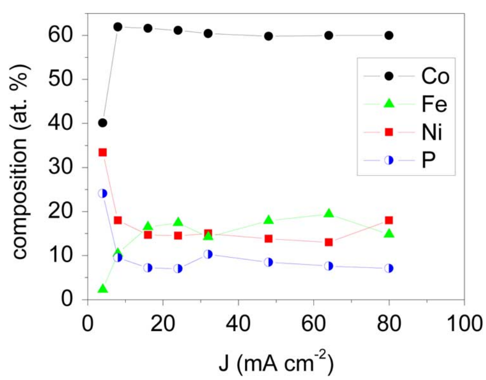


Fig. 1. Composition dependence on plating current density for electrodeposited CoNiFeP films.

increasing coercivity. [7] We suggest a similar mechanism in CoNiFeP. Additionally, at large current densities, hydrogen evolution is considerably higher and the incorporation of hydroxides to the film is likely. This may explain the reduction of the saturation magnetization of films plated at high current density as hydroxides will dilute the magnetization. CoNiFeP are isotropic within the range of current densities studied.

The effect of $NaH_2PO_2$ concentration in the solution on the magnetic properties of CoNiFeP was also studied (data not shown). Films were electroplated at 30 mA cm$^{-2}$ with $NaH_2PO_2$ varying from 0 to 25 mM. The coercivity and magnetization saturation were only sensitive to bath composition between 0 and 5 mM of $NaH_2PO_2$ and remained almost ($\sim$1.5 T) constant for value of concentration between 5 and 25 mM.

As can be seen on the micrograph in Fig. 3, the morphology of CoNiFeP films shows a structure with microcracks. The islands bounded by microcracks are irregular with lateral dimensions varying from 10 to 50 $\mu$m. This behavior is found for all electroplated CoNiFeP films and it does not depend on plating current density or $NaH_2PO_2$ concentration in solution. We attributed the fractures in the films to the stress, which is found in other alloys containing P, such as CoPtP [8].

In a permeability measurement, the relative magnetic permeability $\mu_r = \mu_r' + j\,\mu_r''$ has two components: real $(\mu_r')$ and complex $(\mu_r'')$ parts. The real part of permeability is associated with

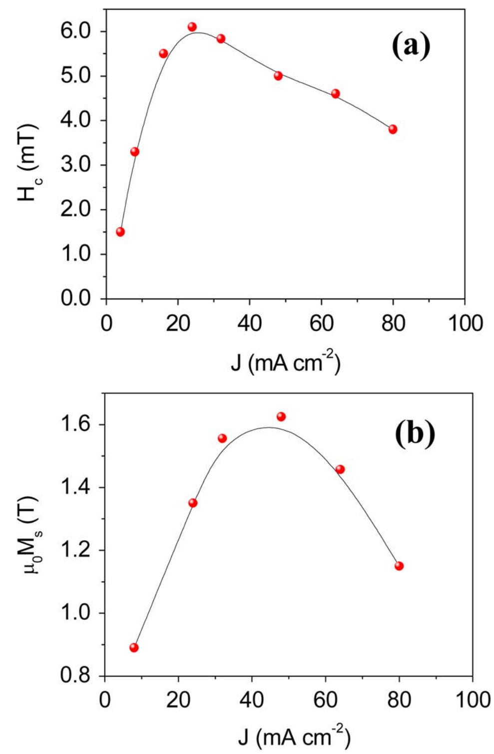


Fig. 2. Dependence of (a) coercivity and (b) magnetization on plating current density at room temperature for electrodeposited CoNiFeP.

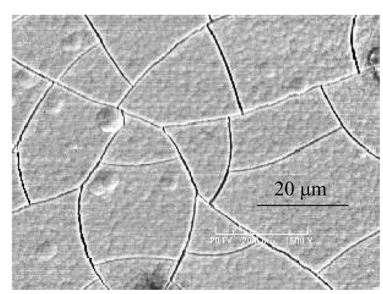


Fig. 3. Top view of an electrodeposited CoNiFeP films showing cracks due to stress.

nondissipative processes, whereas the imaginary part of permeability $\mu_r''$ represents irreversible processes associated with dissipation via Joule effect and/or radio frequency radiation. The dynamic response of a magnetic material to an alternating exciting magnetic field (ac) is usually modeled as an inductor $(L_s)$ in series with a resistance $(R_s)$, where the resistance represents the irreversible processes related to the core losses in the material, which may be frequency dependent. The inductance and resistance are correlated with the real and imaginary part of permeability via [9]

$$\omega L_s = \omega L_0 \mu_r' \tag{1}$$

$$R_s = \omega L_0 \mu_r''. \tag{2}$$

Here, $\omega = 2\pi f$, $L_0$ is the inductance in the absence of a magnetic core, i.e., an air core inductor $(\mu = \mu_0)$. In this low signal

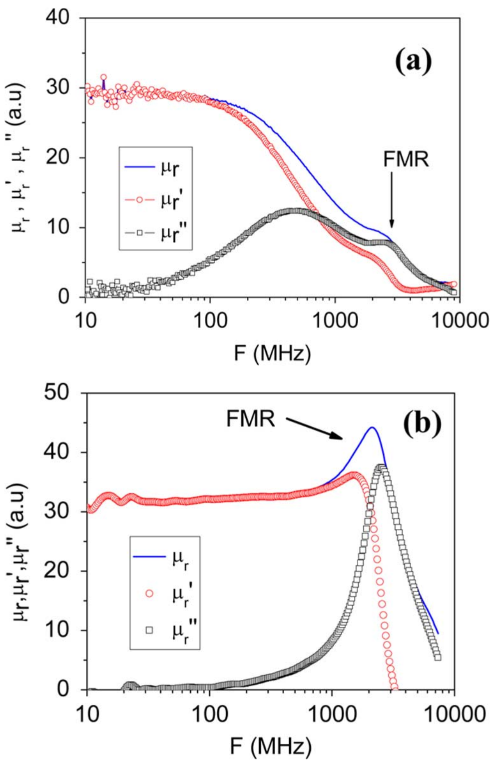


Fig. 4. Permeability spectra of electrodeposited CoNiFeP films with thickness of (a) 28 $\mu$m and (b) 10 $\mu$m.

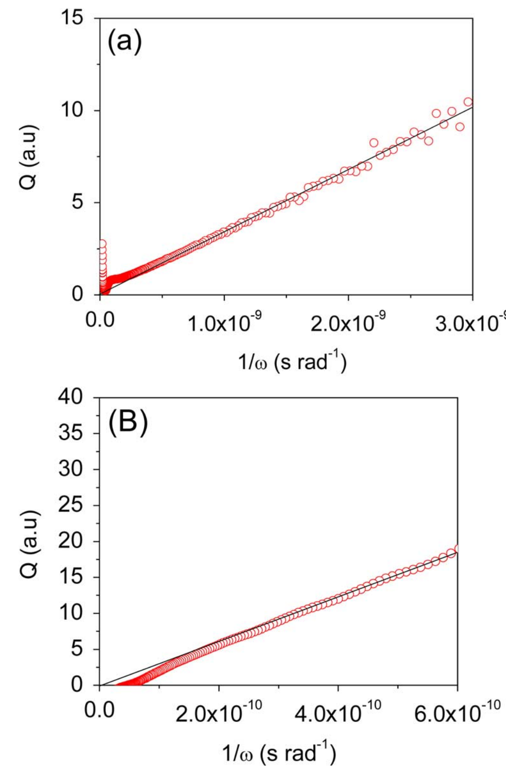


Fig. 5. Quality factor $Q$ versus $1/\omega$ for (a) a 28-$\mu$m-thick and (b) a 10-$\mu$m-thick CoNiFeP film. A linear fitting is plotted as a solid line.

linear model, hysteresis losses are represented by a complex permeability, which is frequency independent, $\mu_r''(f) = \text{constant}$, and $R_s = c\omega L_0$. Dissipation due to eddy current is represented as a linear dependence of complex permeability on frequency, thereby $R_s = \gamma\omega^2 L_0$. The rate of energy dissipated associated with classical eddy current can be calculated from the following:

$$\mathrm{P_{eddy}} = (1/4\rho\alpha)(\omega B d)^2. \tag{3}$$

Here, $\mathrm{P_{eddy}}$ is a rate of dissipated (power loss) per unit volume (W m$^{-3}$), $\rho$ is the resistivity ($\Omega$ m), $\alpha$ is the geometry factor (six for films), $B$ is the magnetic flux density (T), $\omega$ is the angular frequency (rad/s), and $d$ is the film thickness.

Fig. 4 shows permeability spectra of CoNiFeP with thickness of 28 $\mu$m [Fig. 4(a)] and 10 $\mu$m [Fig. 4(b)]. Thickness was varied by controlling the plating time. As the thickness of the films is increased from 10 to 28 $\mu$m, the frequency performance of the magnetic core is reduced. In the case of a 10-$\mu$m sample, the dominating loss mechanism is ferromagnetic resonance, whereas for thick films, it is eddy current.

The quality factor $(Q)$ defined as the ratio between magnetic energy stored and dissipated has a simple relationship when only losses due to eddy current are involved

$$Q = (\rho\alpha/\mu\pi d^2)(1/\omega). \tag{4}$$

The fact that CoNiFeP films have cracks spread over the surface makes it virtually impossible to directly measure resistivity using a conventional four-probe method. During measurement the probing current goes vertically though a CoNiFeP island formed by the fractures and only substrate resistivity may be measured. An alternative approach for direct measurement of resistivity is to pattern the films on a scale smaller than that of the islands and assemble four contacts for electrical measurements. We took an indirect approach to overcome the issue with resistivity measurement by determining it from a permeability spectrum. A similar method has been reported by Ramos *et al.* [10] in hollow circular-cylinder geometry. In this case, the slope of a plot of quality factor $(Q = \mu_r'/\mu_r'')$ versus $\omega^{-1}$ brings information about resistivity according to (4). This equation shows that a linear dependence between $Q$ and $1/\omega$ exists as long as the contribution to the imaginary permeability $(\mu_r'')$ is only eddy current.

Fig. 5 shows the dependence of the quality factor $Q$ on the reciprocal angular frequency $(1/\omega)$. The linear dependence of $Q$ on frequency is a clear indication that eddy current is present and is a significant dissipation mechanism in CoNiFeP thick films. Some deviation from a straight line is observed at low values of $1/\omega$ due to a ferromagnetic contribution. The ferromagnetic resonance frequency (FMR) of CoNiFeP is about 3 GHz as shown in Fig. 4. Therefore, by using the slope of 3.37 $\times$ 10$^9$ rad/s extracted from the linear fitting of Fig. 5(a), $\mu_r =$ 30 from Fig. 4(a), $d = 28$ $\mu$m, we obtain a resistivity of 5.21 $\times$ 10$^{-5}$ $\Omega$ m. Similar value is found for a 10-$\mu$m-thick sample.

Although quality factor can be used as figure of merit, it is not an intuitive parameter for deciding on the size of a magnetic core for an inductor. An alternative figure of merit for inductors is the product of maximum magnetic flux density and operating frequency (BF) at a fixed power loss. Parametric curves with core loss of 500 kW m$^{-3}$ are commonly used to determine a cross section of inductor. For instance, the best ferrite-based inductor has a $\mathrm{BF} = 8 \times 10^4$ T s$^{-1}$. For CoNiFeP, we obtain a $\mathrm{BF} \sim 4 \times 10^5$ T s$^{-1}$ by using (3) with $\rho = 5.2 \times 10^{-5}\ \Omega$ m, $d = 10\ \mu$m, $\alpha = 6$, and $\mathrm{P_{eddy}} = 500$ kW m$^{-3}$. Our BF value is only valid in the limit of high frequency and low $B$ values, where hysteresis loss can be neglected. Therefore, CoNiFeP with BF of $\sim 0.4 \times 10^6$ T s$^{-1}$ may find application in inductors operating at gigahertz frequency range.

## IV. Conclusion

We have studied the influence of addition of P and of plating current density on static and dynamic magnetic and electrical properties of CoNiFe high moment alloy. We prepared films with P content up to 10 at. % and found that it alters film morphology and electrical and magnetic properties. Film composition is weakly dependent on plating current density.

CoNiFeP shows a morphology composed by isolated islands with sizes varying between 10 and 50 $\mu$m. The addition of P increases coercivity up to 6400 A m$^{-1}$ and reduces magnetization saturation down to 1.0 T.

The most remarkable effect of P addition to CoNiFe is the increase of resistivity over two orders of magnitude. The resistivity obtained from permeability measurements is of the order of $10^{-5}\ \Omega$ m compared to $2.4 \times 10^{-7}\ \Omega$ m of pure CoNiFe. The ferromagnetic resonance of CoNiFeP alloy is 3 GHz. This material may find application on high-frequency low signal devices operating in the microwave frequency range.

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Manuscript received February 22, 2008. Current version published December 17, 2008. Corresponding author: S. Roy (e-mail: saibal.roy@tyndall.ie).