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Giant magnetoimpedance in electrodeposited CoNiFe/Cu wire: A study on thickness dependence

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ABSTRACT

Co rich CoNiFe thin films of varying thickness were electrodeposited onto 100 μm diameter copper wires. The microstructure, magnetic properties and magnetoimpedance effect of the samples have been investigated as a function of deposition time. All samples show ferromagnetic behavior with no significant change in coercivity values. The samples are found to exhibit higher magnetoimpedance values (80%) at a frequency of 1 MHz. Detailed analysis of high frequency magnetoimpedance and permeability data suggest a strong correlation between soft magnetic and magnetoimpedance behavior. Following these trends a phenomenological model has also been developed to explain the variation of magnetoimpedance as a function of film thickness.

1. Introduction

Giant magnetoimpedance (GMI), that refers to the change in impedance of a magnetic element supplied with an a.c. input signal as a function of applied longitudinal d.c. magnetic field, has attracted considerable attention recently [1–3]. Studies on GMI phenomenon at lower applied magnetic field is of particular importance for application as magnetic sensor [4,5]. As a consequence, GMI in softmagnetic materials with shapes, such as, ribbons, thin films, multilayer and glass coated amorphous wires [6–11] has been studied for scientific understanding [12,13] and applications [4,5]. Among them, thin magnetic film coated on thin wires appears to be quite promising for the industrial application as they produce large magnetoimpedance (MI) at low applied fields.

The origin of GMI in soft magnetic thin films coated on a non-magnetic wire lies in change of skin depth which in turn depends on factors such as permeability and directionality of applied external magnetic field in conjunction with high frequency a.c. circumferential field created by a current flowing along the sample. So, application of even a small external magnetic field may be expected to tune the skin depth of the sample resulting in large change in it's impedance in accordance with the relation (valid for intermediate frequencies typically from 10 kHz to 10 MHz) [14].

$$Z \propto \frac{1}{\delta} \approx \sqrt{\mu_t \sigma \omega} \qquad (1)$$

Where Z is a.c. impedance of the sample, δ is skin depth, μ_t is transverse permeability, σ is d.c. conductivity of the sample and ω is frequency of a.c. applied signal.

Several methods such as thermal evaporation, sputtering and electron beam evaporation are well known method for deposition of thin films. However, electrodeposition method has been extensively used for thin film deposition on substrates of cylindrical geometry (wires) as this method has a special significance in achieving homogeneity of the film on cylindrical substrates in sharp contrast to other methods of film deposition. Also, the effective control of film quality is achieved in electrodeposition by varying deposition parameters and using d.c. and pulse method of deposition. Accordingly, electrodeposition has been widely used for coating of magnetically soft alloy film on highly conducting wire. GMI effect was observed on these composite wires [15,16]. One of the critical factors affecting the performance of such sensing wires is the thickness of the magnetic coating layer. MI effects have been investigated theoretically for composite wires. An optimum value of film thickness was found for a given value of total thickness of wire at a particular frequency so that the magnetoimpedance effect is maximum [17]. Experimentally Seet et al. verified and reported optimum NiFe film thickness to get maximum value of MI [18]. On the other hand, Atalay and Atalay [11] showed that higher values of MI can be obtained at larger thicknesses of NiFe film. Jantaratana and Sirisathitkul also have reported effect of coating thickness variation of electrodeposited Co/Cu on magnetoimpedance phenomenon quite beautifully [19]. Effect of other deposition parameters, such as, pH of deposition bath and composition of the bath on MI has also been studied on CoNiFe/Cu wire [20–22]. However, film thickness dependence on MI of CoNiFe

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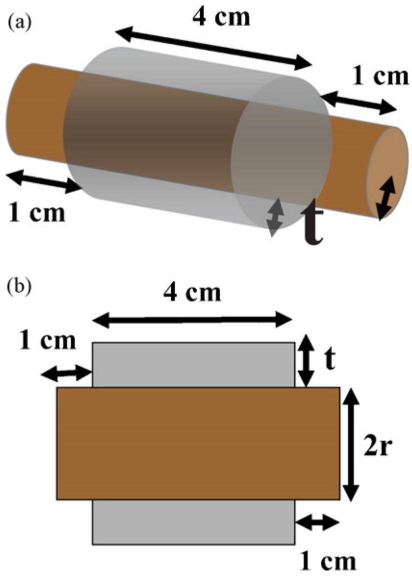

Fig. 1. Schematic diagram of CoNiFe/Cu electrodeposited wire. (a) three dimensional view (b) planner view.

coated on Copper wire has not been investigated. Such studies could be possible if only the thickness of the coated magnetic film is determined. But quite often these measurements are prone to large errors. Alternative approach to determine film thickness, in case of electrodeposited films, appears to be feasible as per the Faraday's law which states that thickness of deposited film varies as

$$h = \frac{Z_E I T_{dep}}{\rho A} \qquad (2)$$

Z_E is electrochemical equivalent of film material, I is current during deposition, T_{dep} is time of deposition, ρ is density of film material and A is area of wire substrate exposed for deposition. In the present work, we have deposited Co rich CoNiFe film on 100 μm Cu wire with varying deposition times. The magnetic and MI properties of the as deposited samples have been investigated. On the basis of present experimental results we propose a phenomenological model to account for the changes observed in MI behavior.

2. Experimental details

All electrochemical experiments were performed in a three-electrode glass cell with a volume of 80 ml, using an electrochemical Analyzer of CH Instruments. All the chemical solutions were freshly prepared by dissolving the requisite amounts of high purity metal sulfates and organic compounds in distilled water. The electrolytic compositions that were used in all the electrodeposition experiments are 60 mM $CoSO_4{\cdot}7H_2O$, 0.1 M $NiSO_4{\cdot}6H_2O$, 5 mM $FeSO_4{\cdot}6H_2O$, 0.2 M H_3BO_3, 10 g/l sodium saccharin and the pH of resulting mixture of solutions was adjusted to 3.0. The working electrode (substrate) was a copper wire of diameter 100 μm, prepared by chemical etching with dilute hydrochloric acid and followed by rinsing in distilled water. Total length of copper wire substrate was 6 cm out of which only 4 cm was exposed for deposition of the magnetic layer. The schematic diagram of the composite wire is shown in Fig. 1 (Fig. 1(a) shows the three dimensional view and Fig. 1(b) shows the planner view). An Ag/AgCl electrode was used as a reference electrode. For all depositions current density (I/A) was fixed at 6 mA/cm^2, Z and ρ being intrinsic property of film material, thickness of film will depend linearly on time of deposition, which was carried out in the range of 30 min to 3 h.

Surface microstructure of the samples has been studied using a Field emission scanning electron microscope (FESEM) of CARL-ZEISS SMT limited, Germany. The quantitative chemical analysis of the alloys were performed by energy dispersive X-ray (EDX). The magnetic characterization of the samples has been done using commercial vibrating sample magnetometer with applied magnetic fields upto 20 kOe, while the frequency dependence of permeability was measured using turn coil method. The impedance was measured in an axial DC magnetic field through the Helmohelz coil, using an Agilent 4294A impedance analyzer with a 42941A impedance probe. The MI data were obtained at a range of frequencies up to 10 MHz, with a constant amplitude a.c. current of 18 mA. The magnetoimpedance percentage is defined as

$$\%\mathrm{MI} = \left(\frac{\Delta Z}{Z}\right)\% = \left[\frac{Z(H_{ext}) - Z(H_{max})}{Z(H_{max})}\right] \times 100 \qquad (3)$$

where H_{ext} is external magnetic field applied, H_{max} is maximum applied magnetic field.

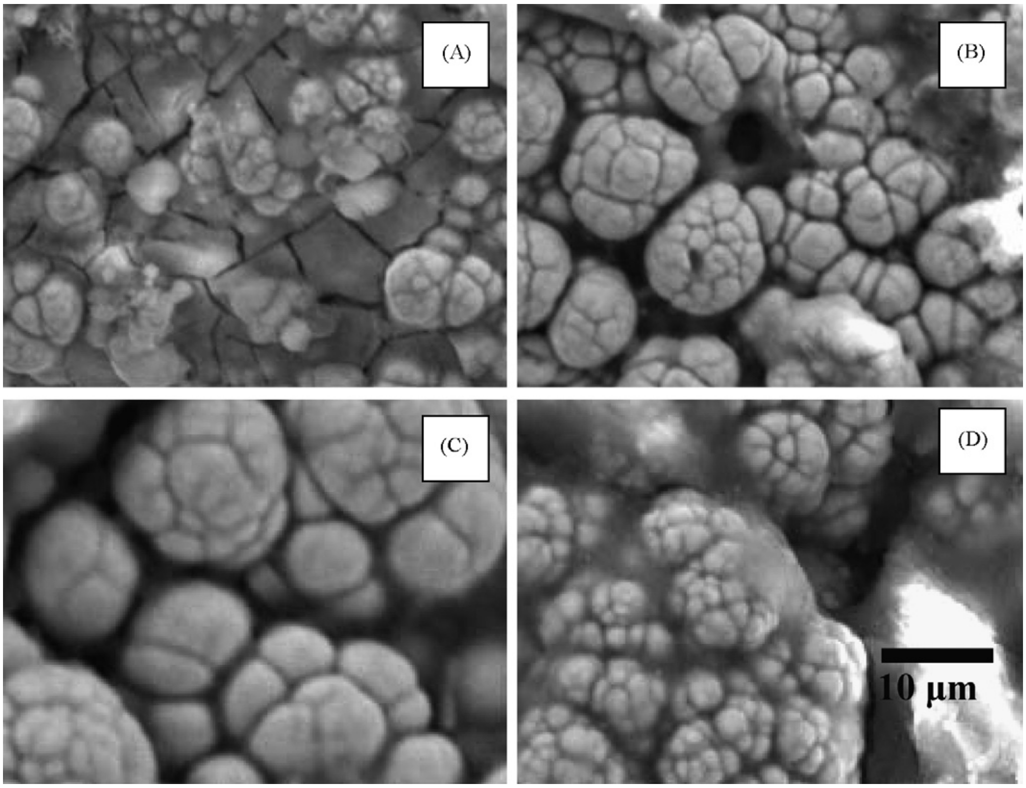

Fig. 2. FESEM micrographs of electrodeposited CoNiFe/Cu wires with deposition time (A) 30 min (B) 60 min (C) 90 min (D) 180 min.

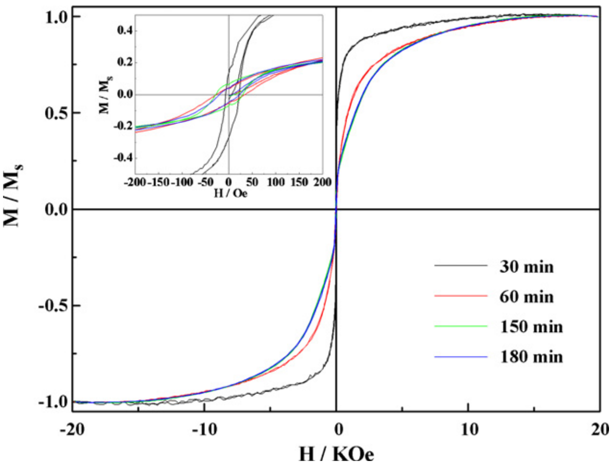

Fig. 3. Field dependence of magnetization at 300 K of samples of three deposition times. Inset: Variation of coercivity with deposition time.

3. Results and discussion

Fig. 2 shows the FESEM surface micrographs of the CoNiFe/Cu wires with varying times of electrodeposition (T_{dep}) i.e, 30, 60, 90 and 180 min. A clear morphological modification has been observed in coated wires as a function of deposition time. It can be seen that the grain sizes in the coated layer are small which develop over the cracked surface in the 30 min deposited wire. Cauliflower-like grain overgrowths were developed on the surface for prolonged deposition times and these self-similar structures seem to increase with increasing in deposition time. The average composition of CoNiFe/Cu for 120 min deposition time was evaluated by EDX microanalysis. This result indicates that the sample has a composition of 75% Co, 5% Fe, and 20% Ni.

Fig. 3 shows magnetic isotherms taken at 300 K under the maximum field of 20 kOe for three typical cases i.e, T_{dep} 30, 60, 150 and 180 min. It can be noticed that the magnetization process is different in 30 min and higher time deposited samples. The magnetization (M) increases sharply on application of low field in 30 min deposited sample compared to others. In all the samples a complete saturation in M could not be attained even at 20 kOe. This could be due to two reasons (i) development of crystalline anisotropy i.e, in the initial stages of deposition it is possible that the Cobalt content could be less. However, at higher T_{dep} the Cobalt content attains the average value determined from EDX, which in turn could lead to higher anisotropy and non-saturation behavior. (ii) The surface oxidation on the thin layer can result in antiferromagnetic or paramagnetic state and that can also result in non saturation M–H curve. Since the H_C values do not vary significantly the reason (i) may not be correct. On the other hand if the oxidation were the main source of these magnetic characteristics, the thin sample would require higher field to saturate. But the results show otherwise.

In electrodeposited samples, during deposition, a current flow along the axis of the wire which produces a circumferential magnetic field and transverse permeability is enhanced, but anisotropy is induced in the sample. The transverse permeability μ_t and impedance Z increase upto the anisotropy field H_K and the decrease as per Eq. (1)

$$Z \propto \frac{1}{\delta} \approx \sqrt{\mu_t \sigma \omega}$$

Fig. 4 shows the field dependence of the MI for electrodeposited wires deposited for three representative times (30, 90 and 180 min) and different frequencies in the range of 100 kHz to 10 MHz. It is observed that the $\Delta Z/Z(\%)$ values initially increase and then decrease on further application of magnetic field H. This double peak behavior was observed in the $\Delta Z/Z(\%)$ vs. H curves of all samples. It is known that the a.c. current flowing across the length of the wire produces a circumferential magnetic field that results in generating a transverse permeability of the ferromagnetic sample is developed.These results show some interesting features:

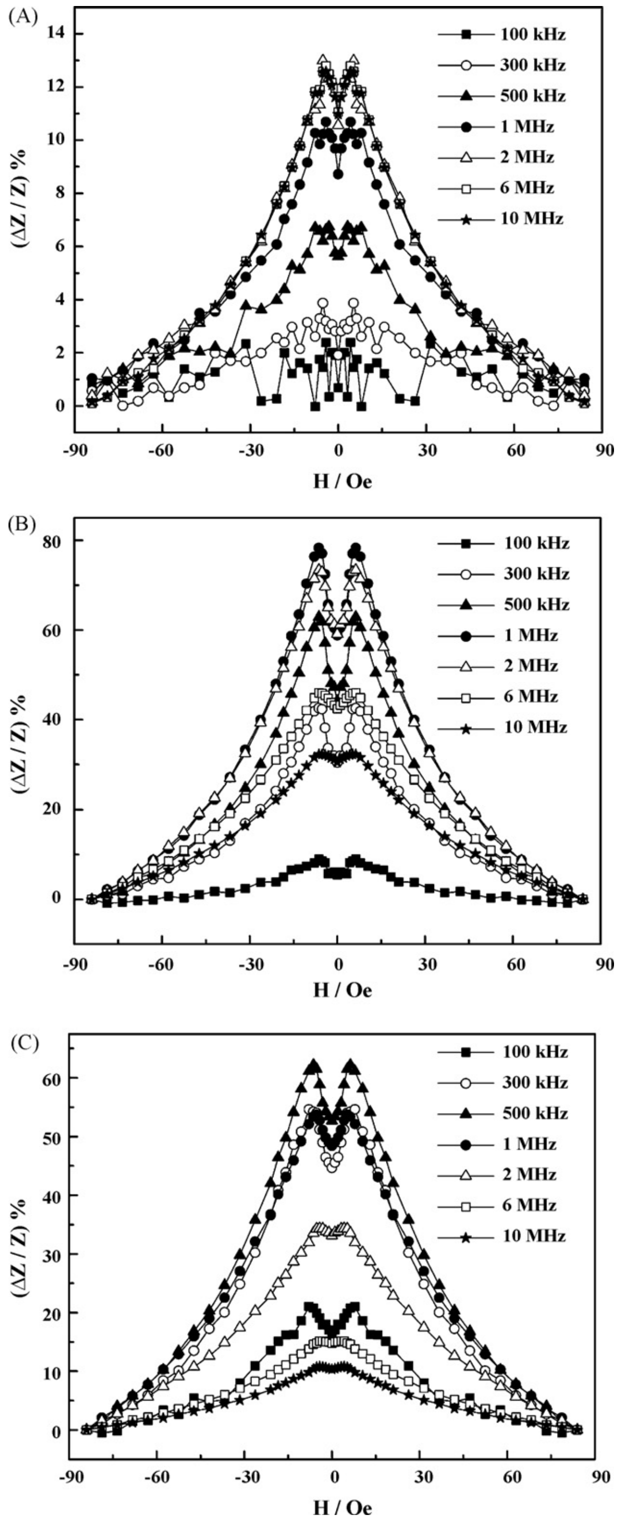

Fig. 4. Magnetoimpedance curves of electrodeposited CoNiFe/Cu wires with deposition time (A) 30 min (B) 90 min (C) 180 min.

(i) In all the cases the MI increases in low fields (~10 Oe) passes through a maximum value $\mathrm{MI}_{\max}$ at a particular field and then decreases at higher applied fields.
(ii) From Fig. 3(A) it can be seen that at lower frequency of 500 kHz in the sample with lower coating thickness, MI appears to be weak and noisy. However, at higher frequencies (>500 kHz) the maximum MI ($(\Delta Z/Z)\%_{\max}$) increases to a value of 13%. This could be attributed to the fact that at low frequency skin depth is more and since magnetic film thickness is less in that particular sample, most of the current flows in Cu core so that change in impedance is not appreciable. However, when the thickness of the film or frequency of the exciting field is increased the dominant contribution from the magnetic film can be seen.
(iii) A $(\Delta Z/Z)\%_{\max}$ value of 12% for 30 min deposited sample at 5 MHz frequency has been observed but increases to 80% for 90 min deposited sample at 1 MHz.
(iv) With increase of thickness or T_{dep}, $(\Delta Z/Z)\%_{\max}$ has been observed at lower frequencies.

In order to bring out the finer features the $(\Delta Z/Z)\%_{\max}$ as a function of thickness at different frequencies is shown in Fig. 5. At a frequency of 2 MHz, $(\Delta Z/Z)\%_{\max}$ initially increases with increase in time of deposition (and hence thickness of film increased) till a critical deposition time of 90 min and later decreases with further increase in time of deposition. These deposition times can be converted to respective thickness of films using Eq. (2). As a rough estimation, since Co is majority content in alloy film, Z for Co is taken (1.0994 g/A h), I/A as 6 mA/cm^2, density of cobalt as 8.86 g/cm^3. Also, assuming current efficiency as 90%, we can relate thickness with time of deposition linearly as: $t(\mu\mathrm{m}) = 6.7T_{\mathrm{dep}}(\mathrm{h})$. Thus when time of deposition varies from 30 min to 3 h, thickness of films varies approximately from 3 to 20 μm.

The trend of maximum MI ratio was probably mainly due to the coating thickness variation. In the absence of an externally applied field, the skin depth in the ferromagnetic layer is lower than its thickness at sufficiently high frequencies [17]. Thus, the alternating current flow is restricted to the outer CoNiFe layer and the resistance (and thus impedance) is sufficiently high. Due to the influence of the externally applied field, the effective transverse permeability decreases, resulting in an increase in the skin depth in the outer CoNiFe layer, which results in decrease of impedance. Thus, limiting the flow of alternating current in the inner Cu core, which results in decrease of resistance of the wire and the ratio increases. Thus, for a given frequency of the alternating current, there is an optimum value of thickness at which the variation of the effective wire impedance is the maximum. Outside this critical range, the $(\Delta Z/Z)\%_{\max}$ reduces drastically. In the case of thicker films, most of the alternating current flows in the outer CoNiFe shell with or without the presence of an applied magnetic field. When it is smaller then, most of the alternating current flows through the inner core even when the maximum reference magnetic field is applied. Both situations result in low $(\Delta Z/Z)\%_{\max}$ ratios. The peak value in $(\Delta Z/Z)\%_{\max}$ is observed at T_{dep} 150 min at 300 kHz frequency while the $(\Delta Z/Z)\%_{\max}$ goes on increasing because in lower frequencies (50 kHz), the skin depth is higher and so the maximum MI is expected at higher thickness as explained above.

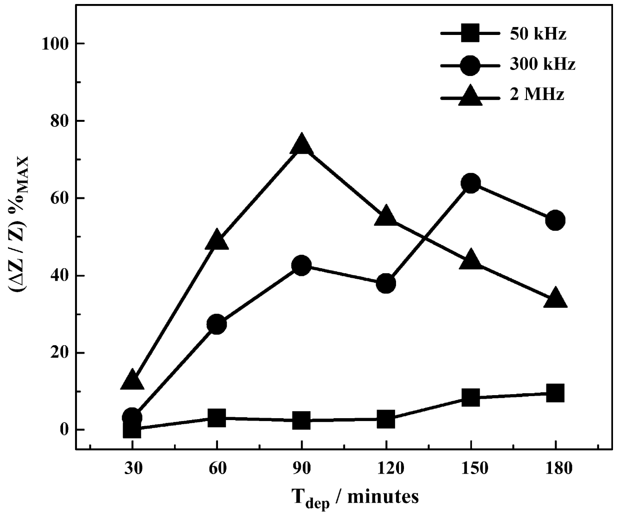

Fig. 5. Maximum Magnetoimpedance vs. time of deposition at different testing frequencies.

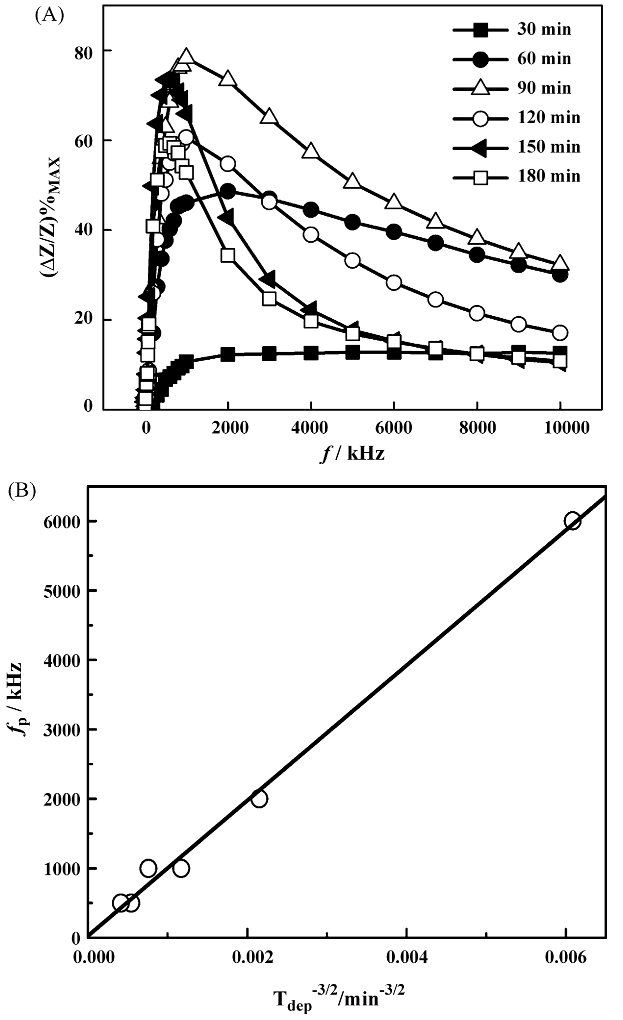

Fig. 6. (A) Maximum Magnetoimpedance vs. testing frequency (B) f_{p} vs. time of deposition

Fig. 6 shows the frequency dependence of $(\Delta Z/Z)\%$ of all the samples of present study. The results indicate that the frequency, at which maximum value of MI occurs, f_{p}, decreases from MHz to kHz frequency range with increasing deposition time (film thickness) which is consistent with the earlier report [11]. As we have discussed, earlier, if skin depth of film material is equal to film thickness then a maximum in $(\Delta Z/Z)\%$ is observed. Since we know skin depth of film material is given as

$$\delta = \sqrt{\frac{1}{\pi \mu_t \sigma f}} \qquad (4)$$

Where μ_t and σ are transverse permeability and conductivity of film material respectively and f is testing frequency.

Following the above arguments we can estimate the value of f_p i.e, the maximum in $(\Delta Z/Z)\%$ is observed when the thickness (t) of the film is equal to the skin depth (δ) ($\delta = t$). If we insert this condition in the Eq. (3) then we can obtain, a relation between t, f_p

$$f_p = \frac{1}{\pi \mu_t \sigma t^2} \tag{5}$$

It follows that with increasing film thickness the peak frequency f_p decreases as t^{-2}. In Fig. 4(B) the thick line represents fitted curve of f_p vs. t which shows $t^{-1.5}$ dependence whereas Eq. (5) predicts it as t^{-2}. This discrepancy can be explained in terms of the frequency dependence of transverse permeability of the film as Eq. (5) does not take an account of it. Earlier Jantaratana and Sirisathitkul have reported $t^{-0.81}$ dependence of f_p for electrodeposited Co/Cu samples. The electrodeposited CoNiFe/Cu samples in present work shows better agreement with theoretical prediction.

Again it can be seen from Fig. 4(A) that as the time of deposition increases, the maximum MI% falls more rapidly with testing frequency. In other words, more the thickness of the magnetic film higher the sensitivity of the sample towards high frequency ranges. To explain this feature, we should note that the fall of MI with frequency is mainly dependent on effective permeability of the sample. More the rate of fall of μ_{eff} with frequency, more rapidly MI will decrease with f.

Now we turn our attention to understand the variation of μ_{eff} with thickness and frequency and attempt to develop a phenomenological model to explain the variation of μ_{eff} with thickness and frequency. Let the uncoated copper wire has conductivity σ_c, permeability μ_0 and the film has conductivity σ_F, permeability μ_F.

Now to determine skin depth of the composite wire, we make a simplified assumption:

The current from top of conductor surface decays as $I = I_0 e^{-\delta x}$. But we assume that the current flow upto the skin depth δ, and is zero afterwards.

Since, the magnetic film is on the top layer, the skin depth as seen by the electromagnetic wave of frequency f is $\delta = \sqrt{\frac{1}{\pi \mu_F \sigma_F f}}$. If the film thickness t is larger than this value of δ for that frequency f, then the copper wire has no role to play. Skin depth of composite wire is then $\delta = \sqrt{\frac{1}{\pi \mu_F \sigma_F f}}$ and effective permeability is μ_F.

However, if t is less than $\delta = \sqrt{\frac{1}{\pi \mu_F \sigma_F f}}$, then extra depth $\sqrt{\frac{1}{\pi \mu_F \sigma_F f}} - t$ has to be covered in copper wire and since the copper wire has different permeability and conductivity, the extra depth penetrated in copper core is enhanced by $\sqrt{\frac{\mu_F \sigma_F}{\mu_0 \sigma_C}}$. Since the wave has also penetrated a distance t in magnetic film before entering copper core, total penetration depth is

$$\delta = \left(\sqrt{\frac{1}{\pi \mu_F \sigma_F f}} - t \right) \sqrt{\frac{\mu_F \sigma_F}{\mu_0 \sigma_C}} + t \tag{6}$$

This is valid till radius of wire is larger than δ which is true in most of the coated wires.

If we assume that σ_{eff} to be the effective conductivity and μ_{eff} as effective permeability of coated wire, then;

$$\delta = \sqrt{\frac{1}{\pi \mu_{\text{eff}} \sigma_{\text{eff}} f}} \tag{7}$$

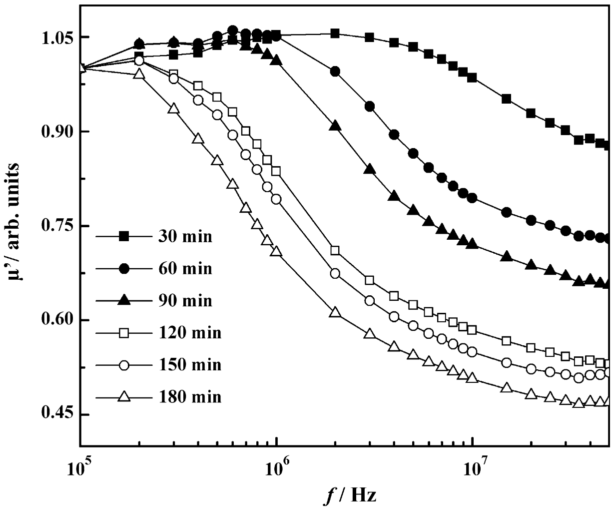

Fig. 7. Longitudinal effective permeability of CoNiFe/Cu coated wires vs. frequency for different thickness.

Equating the two Eqs. (6) and (7) and simplifying, we get;

$$\mu_{\text{eff}} = \frac{(1/\pi f \sigma_{\text{eff}})}{\left[\sqrt{1/\pi \mu_0 \sigma_C f} + t \left(1 - \sqrt{\mu_F \sigma_F / \mu_0 \sigma_C} \right) \right]^2} \tag{8}$$

Taking partial derivative of μ_{eff} w.r.t. f we get,

$$\frac{\partial \mu_{\text{eff}}}{\partial f} = \frac{(1/\pi f^2 \sigma_{\text{eff}}) \left[4\pi^2 f (\partial \mu_F / \partial f) \sqrt{\sigma_F / \mu_0 \sigma_C \mu_F} - 1 + \sqrt{\mu_F \sigma_F / \mu_0 \sigma_C} \right]}{\left[\sqrt{1/\pi \mu_0 \sigma_C f} + t (1 - \sqrt{\mu_F \sigma_F / \mu_0 \sigma_C}) \right]^3} \tag{9}$$

Assuming $\partial \mu_F / \partial f$ behavior remains fairly same for all thicknesses, since ferromagnetic materials have $\mu_F \gg 1$, the second term in denominator is negative and so with increasing film thickness t, $\partial \mu_{\text{eff}} / \partial f$ will increase and this in turn makes the frequency variation of $(\Delta Z/Z)\%$ sharper. Fig. 7 shows the real part of normalized initial permeability vs. frequency plot of all thicknesses where the variation of effective permeability is sharper as the thickness increases which gives a direct evidence of the argument given through Eq. (9).

4. Conclusion

Magnetic and magnetoimpedance bahavior has been investigated on thin films of CoNiFe deposited on 100 μm thick copper wire. These studies have been carried out on the films deposited at various time durations. A maximum value in magnetoimpedance has been observed at an optimum value of film thickness and frequency. The rise and fall of magnetoimpedance with frequency is observed to be much sharper for thicker films. These variations which are strongly related to the permeability of the sample could be understood on the basis of phenomenological model proposed in this work.

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