

Magneto-Impedance Effect of Composite Wires Prepared by Chemical Plating under DC Current

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Abstract: CuBe composite wires of 100 µm in diameter coated with a layer of NiCoP were prepared by a chemical plating method under DC current (CPUDC). The influences of DC current on coating morphology, deposition rate, composition, giant magneto-impedance (GMI) effect and magnetic properties were investigated. It was shown that the circumferential domain structure of coating layer was induced by the DC current going through the wires. A maximum GMI ratio of 870% was obtained in the composite wire prepared under 150 mA and tested at 180 kHz. It is 30 times higher than that of the composite wire plated in the same condition by conventional chemical plating method, indicating that CPUDC is an easy and effective approach to obtain composite wires and its applications will be further extended on magnetic sensors.

Keywords: GMI; Chemical plating; Magnetic properties

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Introduction

Recently, giant magneto-impendence (GMI) effect has been intensively studied for its potential applications in highly sensitive magnetic sensors with high spatial/temporal resolution since it was firstly observed in Co-based amorphous wires in 1992 [1,2]. Experimental and theoretical results confirmed that the origin of the GMI is classical skin effect [4-6]. Although the studies on GMI effect was initially focused in homogeneous materials [1-3], great attention has been paid to the heterogeneous materials such as composite wires and sandwiched films [7,8], since it has great advantages over the homogeneous materials such as [9]: (1) the GMI effect was enhanced obviously, which results from the enhanced skin effect of the coating layer caused by electromagnetic interactions between the conductive and ferromagnetic layers; (2) the MI effect can be observed at much lower frequencies; and (3) realization of miniaturization and practicability for sensors.

Commonly, the composite wires were fabricated by magnetron sputtering [11], electroplating [12-16], chemical plating [17], or MEMS technique [18]. However, the GMI sensitivity of the composite wires is not high since their magnetic structure cannot be well controlled by these methods. It was found that GMI sensitivity could be increased through subsequent processes such as tensile stress annealing treatment [19, 20] and Joule annealing treatment [21].

In this paper, a new technique based on chemical plating was proposed and NiCoP/CuBe composite wires were fabricated. DC current was applied on the wire during the plating process. The influence of pH value of the plating bath and the current magnitude on GMI effect and magnetic structure of the wires were analyzed.

Experimental details

All chemicals used in this work were purchased from

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Sinopharm Chemical Reagent Co., Ltd. (China). Nickelous sulfate (NiSO₄· $6H_2O$), cobalt sulfate (CoSO₄· $7H_2O$), sodium tartaric (Na₂C₄H₄O₆· $2H_2O$), ammonium sulfate ((NH₄)₂SO₄), sodium hypophosphite (NaH₂PO₂· H_2O), stannous chloride dehydrate (SnCl₂· $2H_2O$), palladium chloride (PdCl₂) were used without further purification.

The composite wires were prepared by chemical plating under DC current (CPUDC) along a CuBe wire in diameter of $100 \ \mu m$. Firstly, the wire was rinsed in hydrochloric acid (6 mol/L HCl) for 5 min, followed by sensitization in sensitizing solution (0.11) $mol/L SnCl_2 \cdot 2H_2O$, 0.48 mol/L HCl) for 5 ~ 10 min and activation in activate fluid $(2.8 \times 10^{-4} \text{ mol/L})$ $PdCl_2$, 0.48 mol/L HCl) for 10 min. The plating solution consists of NiSO₄· $6H_2O$ (0.025 mol/L), $CoSO_4 \cdot 7H_2O$ (0.035 mol/L), $Na_3C_6H_5O_7 \cdot H_2O$ (0.10 mol/L), C₄H₄O₆Na₂·2H₂O (0.15 mol/L), (NH₄)₂SO₄ (0.50 mol/L), NaH₂PO₂·H₂O (0.30 mol/L) and different contents of NaOH. After fully mixing the plating solution, the chemical plating process was carried out by the conventional method and CPUDC method, respectively. The temperature of the plating bath was kept at 363 ± 1 K and the pH value was adjusted in the range of $8.0 \sim 8.8$ by adding NaOH dropwise, while the DC current varied from 0 mA to 250 mA.

The morphology and thicknesses of the core and the plated layer were observed using scanning electron microscopy (SEM, RILIs-4800). The composition of the plated layer under each DC current was determined using an energy dispersive X-ray (EDX) device. The hysteresis loops were tested using a Lakeshore 7404 vibrating sample magnetometer (VSM). The magnetoimpedance measurement of the composite wires in length of 4 cm was carried out using a precise impedance analyzer (HP 4294A). The RMS value of alternating current was maintained at 10 mA, and its frequency was varied from 100 Hz to 100 MHz. The applied external magnetic field was generated by a pair of Helmholtz coils and the maximum value of external magnetic field was 100 Oe. During the measurements, the composite wires were subjected along the longitudinal direction and vertical to geomagnetic field.

The GMI ratio was defined as

$$\Delta Z/Z = \frac{Z(H_{ex}) - Z(H_0)}{Z(H_0)} \times 100\%$$
(1)

where $Z(H_{ex})$ and $Z(H_0)$ are the sample impedance values under a magnetic field of H_{ex} and 0 Oe, respectively. The maximum applied magnetic field was 100 Oe.

Results and discussion

Figure 1 shows the GMI effect curves tested at 400 kHz for the composite wires prepared by conventional

chemical plating under different pH values. Initially, the MI ratio increases with H_{ex} to a maximum value, and then decreases with further increase of the field. It means that the easy axis of the coating laver is orientated in circumferential direction [3]. However, the field sensitivity defined as $d(\Delta Z/Z)/dH_{ex}$ is not high, which means that the moment direction deviates from the circumferential direction. When the pH value was increased from 8.0 to 8.8, the maximum *MI* ratio increases from 8.9% to 24.3%, and then decreased to 17.0%. The pH value of the bath may change the composition and magnetic properties of the coating layer. It is evident that the highest *MI* ratio was obtained when pH value is 8.4. The optimal pH value is much higher than that of electroplating method [13-16,22]. The pH value was kept at 8.4 in the following CPUDC method.



Fig. 1 Field dependence of the GMI ratio of the composite wire under different pH values tested at 400 kHz.

Figure 2(a) shows the surface morphology of CuBe wire before deposition. The surface is smooth and uniform. Figure 2 shows the surface morphology of composite wires prepared by conventional chemical plating (Fig. 2(b)) and under DC current of 50 mA (Fig. 2(c)) or 250 mA (Fig. 2(d)). It can be seen that the surface of deposited layer is less smooth and uniform. Evidently, there are tiny holes on the surface of deposited layer because of oxidation-reduction reaction. The number of holes increases with the magnitude of the DC current, giving rise to higher roughness at larger current.

Figure 3 shows the relationship of the deposition rate versus the DC current. It can be seen that the deposition rate increases monotonously with the DC current. The essence of chemical deposition process is oxidation-reduction reaction, associated with the reaction accompanied by electron transfer at the surface of the substrate. With a magnetic field induced by the DC current, the solution near substrate was slightly agitated and the motion of free ions was accelerated because of the microstructure magneto hydrodynamics effect (MHD effect) [22]. Hence, the reaction rate increased by the acceleration of ion motion. This behavior reveals that the deposition rate increases with



Fig. 2 FESEM images of CuBe wire (a) and the composite wires with DC current of 0 mA (b); 50 mA (c); and 250 mA (d).



Fig. 3 Effect of DC current on deposition rate of the coating layer.

the augmentation of the induced magnetic field. Nevertheless, the increased DC current induces higher roughness of the surface during oxidation-reduction reaction, which impedes further augmentation of the deposition rate. Consequently, the magnetic field induced by proper DC current, 100 mA in this work, enhanced the deposition rate significantly due to these two effects.

The composition of the coating layers plated under different DC current was measured by EDX. The current has slight influence on the composition of wires, shown in Fig. 4. Apparently, the variations of the Co, Ni and P percentage in the layers plated under the DC current ranging from 50 mA to 250 mA are all small. The average composition of the coating is $Ni_{27}Co_{59}P_{14}$.



Fig. 4 Effect of DC current on chemical composition of the coating layer.

Figure 5 shows the MI ratio curves tested at 400 kHz for the composite wires plated under different current. It is shown that the MI ratio of the composite wires increases initially and then decreases with the current. A remarkable enhancement of MI ratio occurs at 150 mA. Besides, the field sensitivity of MI curves for the samples with DC current are obviously increased relative to the sample fabricated without current. It reveals that the magnetic domain in these samples are circumferential orientated. Thus, circumferential domain structure in coating layer can be induced by CPUDC method, and the GMI ratio of composite wires prepared using this method improves significantly.



Fig. 5 Field dependence of the GMI ratio of the composite wires prepared by CPUDC method under different DC current.

It is obvious that the maximum GMI ratio increases firstly and then decreases with the increasing of DC current. A maximum GMI ratio of 868% was obtained for the sample deposited under 150 mA. Though the result is a little lower than the ratio of 1100% in electroplated NiFe/Cu composite wire after Joule annealing, CPUDC is one step method [23]. When the DC current is smaller, the induced circumferential structure may increase the GMI ratio. With further increase of the current, the coating roughness and thickness increased. The induced field decreases with the coating thickness layer according to the Ampere law. Consequently, the magnetic moments direction of the magnetic film may orientate from circumferential direction to longitudinal direction [24]. In addition, thicker coating layer results in smaller dynamic driving field, which leads to the weaker magnetization of the magnetic layer. Both the moment deviation and weaker magnetization can result in the significant decrease of the GMI ratio. As for the current of 150 mA, it is the optimal value to get largest GMI ratio.

In previous study [13,22], GMI effect can be enhanced by Joule annealing which induces circumferential magnetic domain. The corresponding annealing current ranges from several hundred milliamperes to several amperes. In this work, CPUDC method could induce circumferential magnetic domain structure during the process of sample preparation. Compared to Joule annealing, this method has simplified the experimental set-up. Meanwhile, the amplitude of the current is smaller.

Figure 6 shows GMI spectra of the samples deposited under different DC current. It can be seen that with frequency increasing, the GMI ratio first increases, undergo a peak, and finally drops down. Each sample has a maximum MI ratio at the critical frequency. With the increase of DC current form 50 mA to 150 mA, the critical frequency decreases from 1.44 MHz to 180 kHz. Further increase of the DC current leads to the increase of critical frequency up to 460 kHz. The critical frequency mainly depended on the softness and thickness of the coating layer [25]. The lowest critical frequency was obtained in the composite wire plated under 150 mA.



Fig. 6 Frequency dependence of the maximum GMI ratio under different DC current.

Figure 7 shows hysteresis loops of the composite wires mentioned above. It can be seen from the loops that the wire prepared by conventional chemical plating method has some circumferential magnetic structure. Whereas the sample prepared by CPUDC method shows that their coating layer has better circumferential magnetic structure when the current amplitude is below 150 mA. Apparently, for composite wire deposited under 150 mA, the coercivity and the energy loss are both smaller.

It is well known that the peak field value in MI curves is the effective anisotropy field of the ferromagnetic layer. Figure 8 shows the dependence of the maximum MI ratio and anisotropy fields acquired from Fig. 5 and 7. It can be seen that the H_k values have the same variation trend as DC current, though the numerical value itself is not completely identical. It can be also seen that the GMI ratio is not coincident with the H_k values. The sample plated under 150 mA has the maximum MI ratio. However its H_k value is not the smallest one. Hence, both of the H_k value and domain structure are important for GMI effect. This result agrees well with the above discussion from the MI curves.

Conclusion

In summary, we have prepared the NiCoP/CuBe composite wires by conventional chemical plating method and CPUDC method. It was found that pH value and DC current had significant effect on GMI effect. The optimal pH value by conventional chemical plating method is 8.4. The GMI ratio was much enhanced using CPUDC method, because of the induced



Fig. 7 DC hysteresis loops of composite wires prepared by conventional and CPUDC methods.



Fig. 8 Dependence of the max GMI ratio and anisotropy field of the composite wires on the DC current.

circumferential domain structure in the coating layer. It was found that GMI ratio reached 870% when the DC current is 150 mA. The established method with DC current is convenient and effective to enhance the MI ratio and could be used to prepare sensing element for magnetic sensors.

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