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Dip coating of silica layer on melt-spun Finemet ribbons: surface morphology and electrical resistivity changes

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ABSTRACT

In this study, melt-spun Finemet ribbons were coated by a thin layer of SiO_2 using dip coating method. Amorphous ribbon prepared by melt spinning method and dip coating were done by using a solution of tetraethylen orthosilicate as a SiO_2 precursor, ethanol as solvent and distilled water for hydrolysis. Different thicknesses of SiO_2 layer, namely 304, 349, 451, 526 and 970 nm were obtained proportional to the number of dipping. Surface morphology and chemical composition of the coatings were analyzed by using Scanning Electron Microscope equipped with an energy dispersive spectroscope. The results clearly verified the presence of Si and O elements and confirmed the presence of silica layer on the surface of all coated ribbons. Microstructure and surface morphology of samples showed a smooth and brittle layer. Electrical resistivity of the samples was measured with a standard four-point probe device. The results confirmed an intense in increase of resistivity. Average value of electrical resistivity for coated samples was around 10^4 Ω-m compared to 10^{-6} Ω-m for Finemet ribbons. Capacity of the samples was evaluated by electronic parameter analyzer device in two different frequencies of 100 kHz and 1000 kHz. Impedance measurements of coated samples in 100 and 1000 kHz showed an increase about 70 and 10 times respectively.

Keywords: *Finemet; Electrical resistivity; SiO_2; Dip coating; Surface morphology.*

1. Introduction

Finemet is an ultra-soft magnetic alloy with a nominal composition of $Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$. Finemet alloy represents high saturation magnetization, small coercivity, and negligible magnetostriction [1, 2]. Due to the high magnetic saturation and low coercivity, they are widely used in transformer cores and induction equipment [3-6].

Since the outset of the original composition in 1989, many attempts have been made to enhance their magnetic properties [7, 8]. Currently, various modifications have led to reducing the eddy current loss and increasing working frequency [9-12]. Among these methods, coating an insulating layer on metallic magnets such as Finemet, which reduced core loss by suppressing eddy current loss as main part of loss especially at high frequency ranges [9, 10]. In this regard, Peng et al. [9] prepared a Finemet/$Ni_{0.5}Zn_{0.5}Fe_2O_4$ core/shell structure. The resulting composite exhibited an enhanced permeability, high saturation magnetization and low core loss. They found that the core loss reduced by increasing resistivity of coated layer. Similar results have been reported by other researchers [10, 11].

Researchers have used many substances and methods in order to coat an insulating layer on

the soft magnetic alloy [13-17]. Zhao et al. [18] fabricated a Fe/SiO_2 laminates with the lateral dimension of a few to several hundred micrometers and sub micrometer thickness. They found out that the operating frequency considerably increased as compared to the parent commercial powders. They also concluded that the amorphous silica coating effectively prevented the eddy current power loss at high frequency. In the other study, Lu et al. [19] synthesized $Fe_{70}Co_{30}/SiO_2$ nanocomposite using co-precipitation method. They reported that the performance of the samples in high frequency was strongly dependent on the SiO_2 content. Increasing the content of SiO_2 as an insulating phase can effectively suppress the eddy-current and drastically shift the cut-off frequency toward high frequency range. In addition, they concluded that the low content of insulating layer was insufficient to suppress the eddy current loss, but a higher amount reduced the saturation. In the other study, Wu et al. [20] synthesized iron-based soft magnetic composites with a silicone coating and reported similar results. In the present work, we applied silica as a high electrical resistivity coating on ultra-soft Finemet alloy by dip coating method. According to our best knowledge, a few researches have studied silica coating on Finemet ribbons up to now.

2. Experimental

Ingot of master alloys were prepared by arc melting under protective argon atmosphere and the actual composition were checked by inductivity-coupled plasma (ICP) analysis. Amorphous ribbons with a cross section of 6 mm× 20 μm were prepared by single roller melt spinning method under protective argon atmosphere with copper wheel speed of 25 m s^{-1}. The structure of amorphous ribbons was identified by X-ray diffraction (STOE D-64295) using Cu-K_α radiation. Chemical composition of ribbons was checked by ICP after both alloying and melt spinning processes.

In order to prepare a suitable silica solution, 9 ml of tetraethylen orthosilicate (TEOS, 98% Aldrich) was mixed with 9 ml of ethanol as solvent. The mixture was stirred at room temperature for 30 minutes. Then, 3 ml of distilled water and a few drops of HNO_3 (65%, Merck) were added to the mixture as hydrolysis agent and catalyst, respectively. The mixture was then stirred for 2 hours. Ribbons were cleaned in an ultrasonic bath for 15 min with acetone before each coating step. Samples were coated by a dip coating device at constant speed of 5 m s^{-1}. Two samples were immersed in the solution for 30 and 60 seconds and then placed in the oven at 100℃. Other samples were plunged for 30 seconds and then coating procedure was repeated up to three times. All samples then placed in the oven at 450℃ for 1 hour with the heating rate of 5℃ min^{-1}.

Surface morphology and chemical composition of the coatings were analyzed by using Scanning Electron Microscope (SEM, VEGA- TESCAN) equipped with an energy dispersive spectroscope (EDS). Scanning electron microscopy was performed at room temperature and 15 kV. Electrical resistivity of the samples was measured with a standard four point probe device. The device was designed based on Jandel company standards, including 4 pins made of tungsten carbide, which were placed 25-50 mm from each other. Capacity of the samples was evaluated by electronic parameter analyzer device (K367 Keithley IV-CV American model) in two different frequencies of 100 kHz and 1 MHz.

3. Results and discussion

XRD pattern of the as quenched sample in Fig. 1 shows only a broad halo peak, which is a characteristic of amorphous state. This confirms that the wheel speed in melt spinning was enough high to achieve a fully amorphous structure.

Surface morphology and quality of the resulting SiO_2 coated layer were analyzed by SEM and the corresponding images for single-layer (1 dipping, 30 s) and four-layer (4 dipping, 4×30 s) samples are presented in Fig. 2. As depicted in the figure the coating was successfully applied and by increasing the number of the dipping the thickness of silica

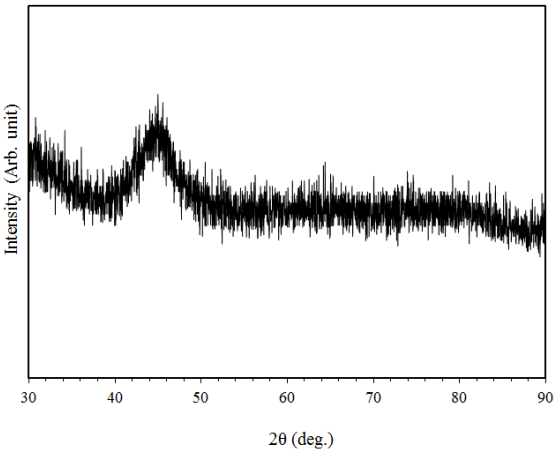

Fig. 1-XRD pattern of as-spun Finemet ribbon at wheel speed of 25 m s^{-1}.

layers (black areas) increases. Some cracks are seen on the surface of coated ribbons. More cracks appeared in the 4 dipping sample (Fig. 2-b) as compared to 1 dipping (Fig. 2-b). The source of these cracks may come from contamination of the solution or substrate due to the environmental impurities. The path of crack propagation as well as microscopic features on the coated surface shows the branched and bifurcated cracks that successively repeated and produced a family of cracks. This show the brittleness of coated layer and especially obvious for multilayer coated sample.

Chemical compositions of the coating layer were also analyzed by EDS and the results are shown in Table 1. The results clearly verified the presence of Si and O elements and confirmed the presence of silica layer on the surface of all coated ribbons. Estimated atomic percent of Si and O, presented in the table, confirms the formation of SiO_2 layer on the ribbons surface. The presence of Au in the

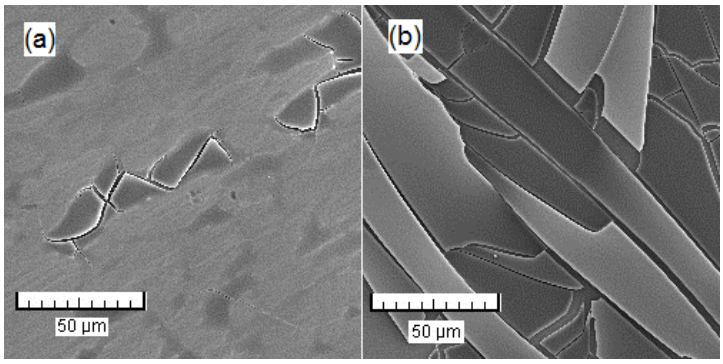

Fig. 2-SEM images of coated ribbons (a) single layer (1 dipping, 30 s) sample (b) four layer (4 dipping, 4×30 s) sample.

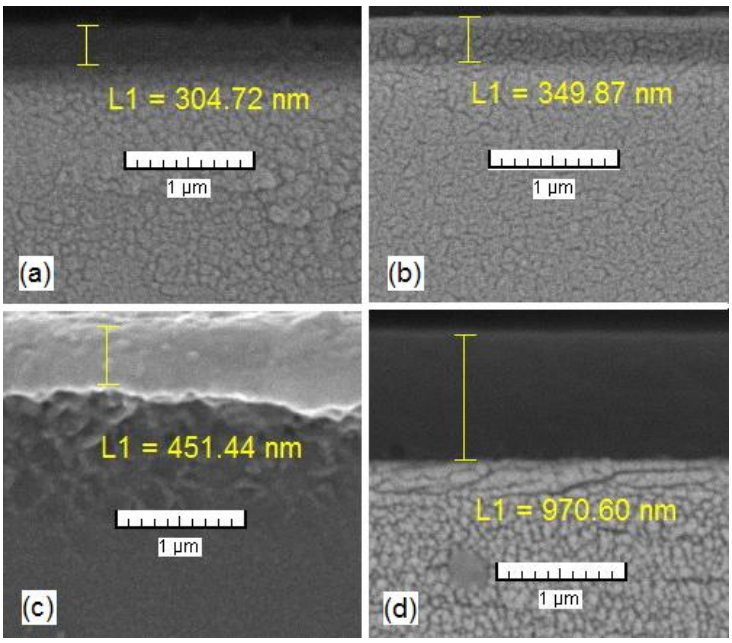

Fig. 3-SEM images of samples (a) single layer (1 dipping, 30 s), (b) single layer (1 dipping, 60 s), (c) two layers (2 dipping, 2×30 s), (d) four layers (4 dipping, 4×30 s).

Table 1- EDS analysis of 4 dipping sample

Element	Line	wt %	at %
O	K_α	57.8	76.6
Si	K_α	29.0	21.9
Au	L_α	13.1	1.4

table relates to Au coating used for preparing the samples.

Fig. 3 shows the cross sectional of SEM images of SiO_2 thin films for different dipping times and cycles. Surface of the coated layer of samples are rather smooth and no observable separation between layers was detected. The high density and high quality of coating confirms that the dip coating is suitable method for preparation of the silica/ Finmet samples. The thicknesses of coating versus dipping number were shown in Fig. 4. As can be seen in the figure the thickness was proportional to the number of dipping.

The relation between dipping time and cycle and the thickness of samples can be understood from Fig. 3 and the results are plotted in Fig. 4. Fig. 3(a) and 3(b) indicate that the increasing of coating time results in a rather slight increase in the layer thickness. Furthermore, a significant increase in the thickness was seen, from 304 nm for the single-layer (1 dipping) to 970 nm for four-layers (4 dipping) coated samples.

Fig. 5 shows the electrical resistivity measurements of amorphous ribbon and the coated samples with different thickness. The resistivity measurements clearly show that the electrical resistivity has a dramatic increase from about 10^{-6} Ω-m for amorphous ribbon to 10^{4} Ω-m for coated

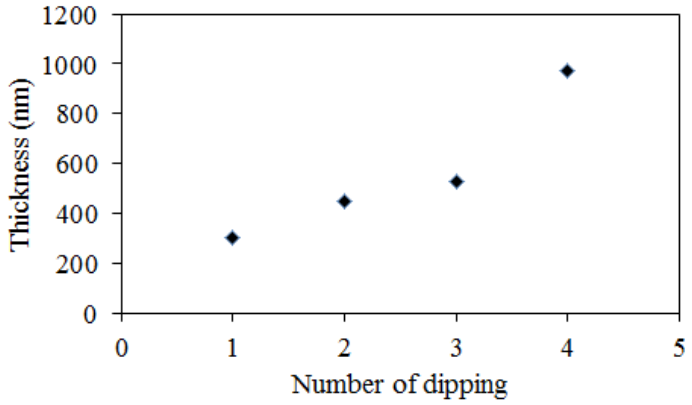

Fig. 4-Plot of coating thickness as function of dipping number.

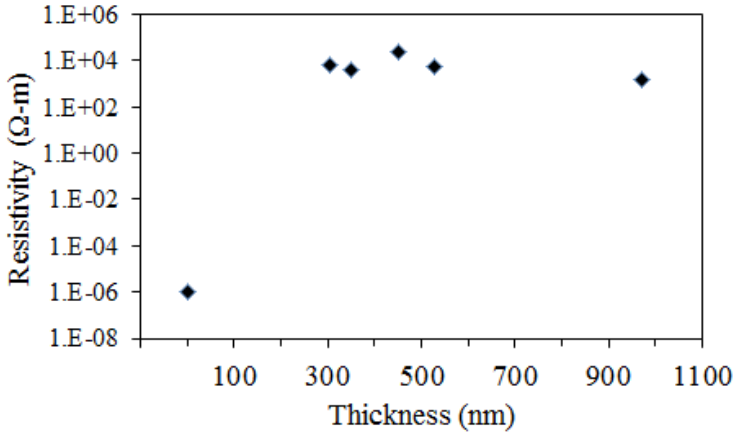

Fig. 5-Electrical resistivity of the amorphous ribbons and coated ribbons with different thickness.

Table 2- Impedance (Z) measurements in 100 kHz and 1 MHz

Sample	Impedance (Ω), 100 KHz	Impedance (Ω), 1 MHz
Substrate	1.0×10^{4}	1.0×10^{5}
1 dipping Coated	7.1×10^{5}	1.4×10^{6}
4 dipping Coated	7.4×10^{5}	1.5×10^{6}

samples. Moreover, the changes of layer thickness do not have a significant effect on the electrical resistivity. Actually, the increase in the electrical resistivity of samples is a direct consequence of the high resistivity of the silica layer. Considering the fact that Finemet ribbons with one SiO_2 layer have approximately the same electrical resistivity in comparison to multilayers samples, therefore, samples produced by even 1 dipping procedure is a good candidate for use in high resistivity and high frequency applications.

Results of the impedance (Z) measurements in 100 kHz and 1 MHz were shown in Table 2. According to these results, impedance had an increase in both frequencies. Furthermore, the number of coated layers on the Finemet has no considerable effect in impedance quantity, especially in 1 MHz.

4. Conclusion

Finemet ribbon with a SiO_2 coating was prepared by dip coating method. SEM micrographs showed that the surface of the samples contained a thin and smooth insulating layer of silica without any separation. Coating thickness was proportional to the number of dipping cycle. Results showed that the silica coating increased the electrical resistivity, as well as impedance.

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