

STRUCTURAL, ELECTRICAL AND MAGNETIC PROPERTIES OF (FeNi)-Zr-Nb-N THIN FILMS

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Nanocrystalline (FeNi)-Zr-Nb-N thin films consisting of a mixture of bcc and fcc phases and a small amount of amorphous phase were deposited in argon (Ar) and nitrogen (N₂) atmosphere, in a sputtering system at room temperature. Some results concerning the effects of composition and thermal treatments on the structural, electrical and soft magnetic properties of (FeNi)-Zr-Nb-N thin films, in view of their utilization for manufacturing magnetic microdevices, which operate at high frequencies, are presented. Among the compositions of investigated thin films, (Fe₈₅Ni₁₅)₈₂Zr₂Nb₄N₁₂ samples exhibit excellent electrical and soft magnetic properties: good electrical resistivity $\rho \cong 125 \mu\Omega\text{cm}$, high saturation magnetization $4\pi M_s \cong 19 \text{ kGs}$, low coercivity $H_c \cong 2 \text{ Oe}$ and near zero – magnetostriction. The good magnetic properties were due to the decrease of FeNi grain size by mixed effect of the intergranular amorphous phase as well as Zr and Nb – based precipitates.

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1. Introduction

Soft magnetic nanocrystalline materials are subject of great current interest because of their promising technological applications. In recent years, the micromagnetic devices, such as magnetic thin film inductors, have been developed to miniaturize the magnetic components in electronic equipments [1-5]. The materials used for manufacturing magnetic thin films inductors need to present specific characteristics: high saturation magnetization ($4\pi M_s$), low coercivity (H_c), high effective permeability, low magnetostriction, controllable anisotropy, good thermal and temporal stability of magnetic properties, high corrosion resistance and adherence to substrate [2]. It is well known that FeNi alloy thin film is very important soft magnetic materials due to their high magnetic moment and low magnetostriction. Its magnetic and electrical characteristics, however, often make the material too lossy to be used at above $\sim 100 \text{ MHz}$. The (FeNi)-M-N thin films, with (M = Nb, Zr) exhibit very good soft magnetic properties attributed to nanocrystalline nitrides (Fe,Ni)-N phases. Concurrently, high electrical resistivity of (FeNi)-M-N magnetic thin films receive much attention from the standpoint of low core-loss for high frequency use.

This paper reports some results concerning the effects of composition (Ni concentration and Zr/Nb ratio) and thermal treatment on the structural and soft magnetic properties of (FeNi)-Zr-Nb-N thin films, in view of their utilization for manufacturing magnetic microdevices which operate at high frequencies. We expect to obtain near zero - magnetostriction alloys by mixing Zr and Nb to the (FeNi) – N films. To improve the soft magnetic properties of (FeNi)-M-N thin films, thermal treatments have been performed.

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2. Experimental

The (FeNi)-Zr-Nb-N thin films were prepared using a rf diode – sputtering system (Laboratory Sputtering Leybold – Heraeus Plant Z – 400). The thin films were deposited at room temperature, in an Ar and N₂ gas mixture, using composite sputtering targets. The partial pressure of N₂ gas was controlled with a spectrometer and during film deposition is maintained at about 8 % of total pressure.

The sputtering targets consisted of FeNi (Fe 95.85 at.% - Ni 5.15 at.%) alloy disks (75 mm in diameter) with small pieces of Zr and Nb on its surface. The film composition was varied by changing the number of Zr and Nb pieces on FeNi alloy targets. The film thickness was about 1.2 μm for all the samples.

For electrical and magnetic measurements the (FeNi)-Zr-Nb-N thin films were deposited on glass substrates. The electrical resistivity of the (FeNi)-Zr-Nb-N thin films was measured using the D.C. four – point probe technique. The magnetic characteristics ($4\pi M_s$ and H_c) of the samples were measured using a vibrating sample magnetometer.

The structure of the (FeNi)-Zr-Nb-N thin films was investigated using X-ray diffraction analysis. An X-ray diffractometer with a monochromatized Mo-K α radiation was used, in a Bragg-Brentano arrangement. The microstructure of the (FeNi)-Zr-Nb-N thin films was investigated by transmission electron microscopy (TEM), using molybdenum ‘microscope grids’, coated with an evaporated carbon (8 – 10 nm) thin film as substrates.

For magnetostriction measurements the (FeNi)-Zr-Nb-N thin films of rectangular shape ($0.1 \times 5 \text{ cm}^2$) were deposited on silicon substrates. Magnetostriction measurements were performed in a magnetic field up to 7.2 kOe using a set-up based on the two-beam free-sample deflection method.

3. Results and discussion

The soft magnetic properties and structure of (FeNi)-N thin films are influenced by Ni and N concentrations. In Table 1 are comparative presented the magnetic and electrical characteristics for as-deposited Fe-N and (FeNi)-N at different Ni concentrations when the N concentration is maintained at 12 at. %.

Table 1. Magnetic and electrical characteristics for as-deposited Fe-N and (FeNi)-N thin films.

Material	Resistivity ($\mu\Omega \text{ cm}$)	Magnetization $4\pi M_s$ (kGs)	Coercivity (Oe)
Fe ₈₈ N ₁₂	105	14	8.5
(Fe ₉₅ Ni ₅) ₈₈ N ₁₂	102	14.9	9.3
(Fe ₈₅ Ni ₁₅) ₈₈ N ₁₂	104	16	10
(Fe ₇₀ Ni ₃₀) ₈₈ N ₁₂	102	16.3	20.5

The as – deposited (FeNi)-N thin films with 15 at. % Ni and 12 at. % N concentrations showed optimal magnetic properties: high saturation magnetization of about 16 kGs and coercivity of about 10 Oe. Concurrently, the magnetic properties of (FeNi)-Zr-Nb-N thin films are strongly affected by the Zr+Nb amount and the Zr/Nb ratio. For studying the influence of the Zr and Nb concentrations on the magnetic properties, the Ni and N contents in (Fe₈₅Ni₁₅)_xZr_yNb_zN₁₂ thin films are maintained at 15 at.% and 12 at.%, respectively. The magnetic properties for (Fe₈₅Ni₁₅)_xZr_yNb_zN₁₂ thin films were investigated by choosing the Zr and Nb as components in ratio of 1/2, for different values of y and z, respectively (i.e. 1+2; 2+4; 3+6; 4+8 and 5+10 at.%).

The dependence of the saturation magnetization $4\pi M_s$ and coercivity H_c on the Zr and Nb amounts ($y+z$) for (Fe₈₅Ni₁₅)_xZr_yNb_zN₁₂ thin films is presented in Fig. 1. One can be see that the values of saturation magnetization decrease with increasing the Zr and Nb content up to 4 and 8 at. %, respectively and slowly increase afterwards. At the same time it can be observed that the coercivity values have a minimum for Zr and Nb content around 2 and 4 at. %, respectively.

By analyzing the presented results it can be seen that the best soft magnetic properties have been obtained for Zr+Nb = 2+4 = 6 at. %. In the same time the electrical resistivity value for as-deposited (Fe₈₅Ni₁₅)₈₂Zr₂Nb₄N₁₂ thin films is about 150 μΩ cm.

The magnetic measurements and structural analysis were made on as-deposited and thermally treated thin films. The $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films were successively thermally treated, in vacuum, for 1 h, at temperatures between 200 and 600 °C.

The dependence of saturation magnetostriction on Zr and Nb amounts ($z+y$) for $(\text{Fe}_{85}\text{Ni}_{15})_x\text{Zr}_y\text{Nb}_z\text{N}_{12}$ thin films is presented in Fig. 2. One can see that for Zr and Nb content of 2 and 4 at. %, respectively the near zero – saturation magnetostriction values are obtained.

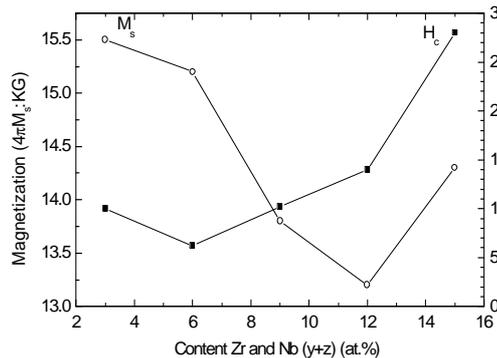


Fig. 1. The dependence of the saturation magnetization and coercivity on Zr and Nb content.

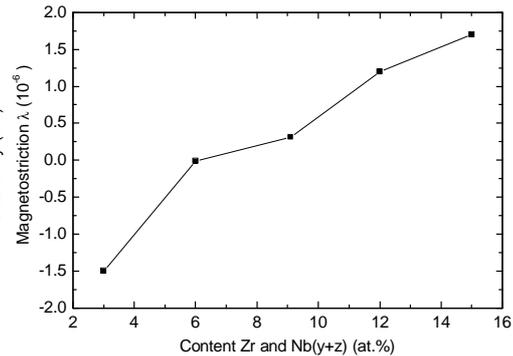


Fig. 2. The dependence of the saturation magnetostriction on Zr and Nb content.

The changes of the magnetic characteristics of samples after successively anneals were studied. The dependence of saturation magnetization and coercivity on annealing temperature for $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films is presented in Fig. 3. One can see the value of the saturation magnetization increases with increasing the annealing temperature. The saturation magnetization values are small for thin films annealed below 400 °C, but increase with increasing annealing temperature, presenting a maximum of 18.5 kGs after annealing at 500 °C. Simultaneously, the coercivity values reached a minimum of about 2 Oe, after annealing at 500 °C.

In Fig. 4 are presented the X-ray diffraction patterns for $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films in as-deposited state and after annealing at 400 °C and 500 °C. The X-ray diffraction patterns of the $(\text{Fe}_{85}\text{Ni}_{15})\text{-N}$ films have revealed only a halo specific for amorphous state.

The X-ray diffraction patterns of as-deposited $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films reveal a structure, which consists of amorphous and nanocrystalline phases (mixture of bcc and fcc FeNi). This mixed structure was also confirmed by the temperature dependence of the saturation magnetization, by identifying the corresponding Curie temperatures.

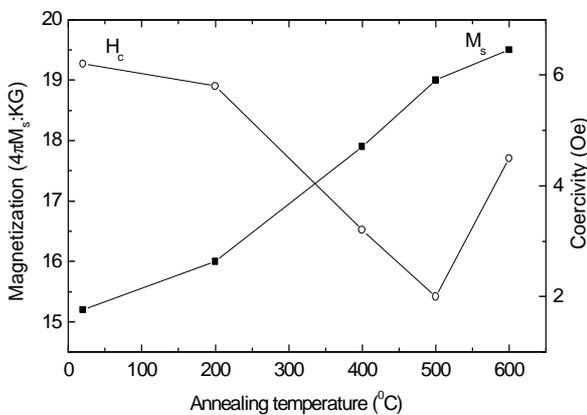


Fig. 3. The dependence of magnetic characteristics on the annealing temperature for $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films.

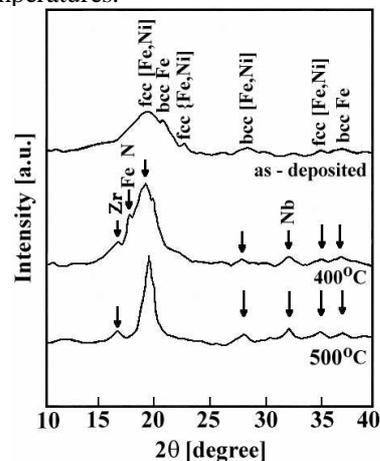


Fig. 4. X-ray diffraction patterns for $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films.

The fraction of the crystalline phase increased with the increase of the annealing temperature. The structure of annealed $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films shows the coexistence of a mixed bcc and fcc FeNi crystalline phase and a residual amorphous phase.

After annealing at 400 °C and 500°C, the X-ray diffraction patterns of $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films present a peak at $2\theta = 19.5$ deg., which corresponds to the bcc (Fe,Ni) reflections and two small peaks at $2\theta = 28$ deg. and $2\theta = 35$ deg., which correspond to the bcc and fcc (Fe,Ni) reflections. Distinct weak lines of Zr and Nb phases arise in X-ray diffraction patterns after annealing. The structural changes produced after annealing at 400 and 500°C are also confirmed by changes in graphs of the Fig. 3 representing the dependence of soft magnetic characteristics on the annealing temperature.

The changes of the electrical resistivity after successively annealing were also studied. As compared to as-deposited $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films, the annealed samples present smallest values of the electrical resistivity (about 125 $\mu\Omega\text{cm}$) due to the recrystallization process and probably a decrease in the nitrogen concentration in film.

TEM micrographs of $(\text{Fe}_{85}\text{Ni}_{15})_x\text{Zr}_y\text{Nb}_z\text{N}_{12}$ film for two different compositions are shown in Fig. 5. The average grain size is large (about 50 nm in diameter) for $(\text{Fe}_{85}\text{Ni}_{15})_{85}\text{Zr}_1\text{Nb}_2\text{N}_{12}$ film, but decreases at about 15 nm when the Zr and Nb amounts (y+z) increase at 6 at.%, for $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ film.

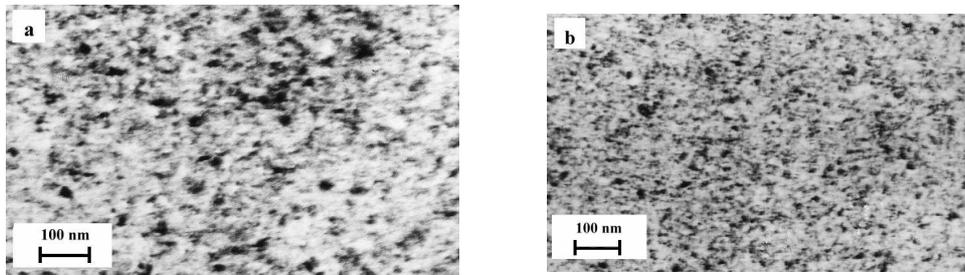


Fig. 5. TEM micrographs of $(\text{Fe}_{85}\text{Ni}_{15})_{85}\text{Zr}_1\text{Nb}_2\text{N}_{12}$ film (a) and $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ film (b).

As compared to $(\text{Fe}_{85}\text{Ni}_{15})_{88}\text{N}_{12}$ thin films which are widely used for magnetic microdevices, the $(\text{Fe}_{85}\text{Ni}_{15})_x\text{Zr}_y\text{Nb}_z\text{N}_{12}$ thin films showed good soft magnetic properties due to the decrease of the grain sizes of (FeNi) nanocrystalline phase.

4. Conclusions

Soft magnetic $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films with high saturation magnetization $4\pi\text{Ms}$ of about 19 kGs, high electrical resistivity of about 125 $\mu\Omega\text{cm}$ and low H_c of 2 Oe have been prepared. The low H_c is due to the grain size reduction at nanometric scale and that is attributed to the grain growth constriction effect of the Zr and Nb nitride nanoparticles that are formed during the reactive sputter deposition. An enhancement of soft magnetic properties for (FeNi)-N system were obtained by adding 2 at. % Zr and 4 at. % Nb, respectively. The good magnetic properties were due to the decrease of FeNi grain size by mixed effect of the intergranular amorphous phase as well as Zr and Nb-based precipitates.

By analyzing the presented results one concludes that $(\text{Fe}_{85}\text{Ni}_{15})_{82}\text{Zr}_2\text{Nb}_4\text{N}_{12}$ thin films are interesting for micromagnetic devices including magnetic inductors, which can operate at high frequency.

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