

ON THE GALVANOMAGNETIC AND STRUCTURAL PROPERTIES OF GRANULAR $(\text{CoNi})_x\text{-(SiO}_2\text{)}_{1-x}$ THIN FILMS

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The aim of this paper is to present some results concerning the galvanomagnetic, electrical and structural properties of $(\text{CoNi})_x\text{-(SiO}_2\text{)}_{1-x}$ (for x values between 0.4 and 0.6) thin films in view of their utilization for manufacturing galvanomagnetic devices. The samples were thermally treated at in vacuum at temperatures between 150°C and 300°C, in order to stabilise their physical properties. Their microstructure and composition have been investigated by transmission electron microscopy and X-ray diffractometry, respectively. The galvanomagnetic measurements were made in air, at room temperature, by van der Pauw method. The influence of composition and post – deposition annealing on the microstructure, electrical resistivity and galvanomagnetic sensitivity of $(\text{CoNi})_x\text{-(SiO}_2\text{)}_{1-x}$ thin films is discussed.

Keywords: Thin films, Hall voltage, Galvanomagnetic properties

1. Introduction

Granular metal films with nanocrystalline or amorphous structure are of great interest for applications in the field of miniature devices. Granular metal thin films are metal - metal or metal - insulator mixtures consisting of grains with nanometric size. Recently, interesting galvanomagnetic properties were discovered in magnetic metal-insulator granular thin films [1-2].

The physical properties of the metal - insulator granular thin films are strongly dependent on composition and microstructure. This fact allows the optimisation of the characteristics required for different kinds of devices based on granular films. The metal – insulator thin films with ultrafine structure displays unusual structural, electrical and magnetic properties for values of the metal content near a critical value called percolation threshold. For a wide variety of metal – insulator films the values of percolation threshold have been 0.5-0.6 [3].

The thin film materials used for manufacturing Hall – effect devices need to present specific characteristics: large Hall sensitivity, low linearity error, good temporal stability, good corrosion resistance and adherence to the substrate.

We present some results concerning the electrical, structural and galvanomagnetic properties of $(\text{CoNi})_x\text{-(SiO}_2\text{)}_{1-x}$ (for x values between 0.4 and 0.6) thin films in view of their utilisation for manufacturing Hall – effect devices. The dependence of the structural properties, electrical resistivity and Hall sensitivity on the metal content and annealing temperature of these thin films were investigated.

2. Experimental

$(\text{CoNi})_x\text{-(SiO}_2\text{)}_{1-x}$ (for x values between 0.4 and 0.6) thin films were prepared by R.F sputtering in Ar atmosphere ($p = 10^{-2}$ mbar) using two composite targets as source material. The sputtering targets were made of a CoNi alloy or SiO_2 plate having 7.5 cm in diameter, on which small SiO_2 or CoNi circular pieces (0.5 cm in diameter), were disposed. Modifying the surface ratios of components of the target surface made compositional changes of the samples.

The $(\text{CoNi})_x - (\text{SiO}_2)_{1-x}$ thin films have a thickness of about 350 nm and they deposited at room temperature, on different substrates depending on the intended measurements.

For electrical and galvanomagnetic measurements and X-ray diffraction analysis, the $(\text{CoNi})_x - (\text{SiO}_2)_{1-x}$ thin films were deposited on glass substrates.

The structure of the $(\text{CoNi})_x - (\text{SiO}_2)_{1-x}$ thin films was examined by X-ray diffractometry. An X-ray diffractometer with monochromatized Co $K\alpha$ radiation ($\lambda = 0.178897$ nm) was used in a Bragg–Brentano arrangement.

The microstructure was analyzed by transmission electron microscopy (TEM) and the samples were deposited on standard copper ‘microscope grids’ coated with on evaporated carbon (8-10 nm) thin films. The $(\text{CoNi})_x - (\text{SiO}_2)_{1-x}$ thin films with a thickness of about 60 nm were used for TEM analysis.

The film composition was determined by the electron microprobe analysis on $(\text{CoNi})_x - (\text{SiO}_2)_{1-x}$ thin films deposited on molybdenum substrates.

The electrical resistance of the as-deposited and annealed samples was measured using the standard D.C. four – probe technique.

The galvanomagnetic measurements were made in air, at room temperature, by van der Pauw method, on $(\text{CoNi})_x - (\text{SiO}_2)_{1-x}$ thin films of rectangular shape (0.7×1.4 cm²) [4]. The measurements of the Hall voltage were made for magnetic field induction values up to 2 T.

The structural analysis of the samples and the electrical and galvanomagnetic measurements were made on as-deposited and thermally treated samples. The samples were thermally treated at in vacuum at temperatures between 150°C and 300°C, in order to stabilize their physical properties.

3. Results

We studied the dependence of the CoNi thin film electrical resistivity on the value of the Co content. We also studied the dependence of the $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin film electrical resistivity on the $(\text{Co}_{80}\text{Ni}_{20})$ content. These dependencies, for as-deposited and annealed samples (at 300°C for 1 h) are presented in Fig. 1.

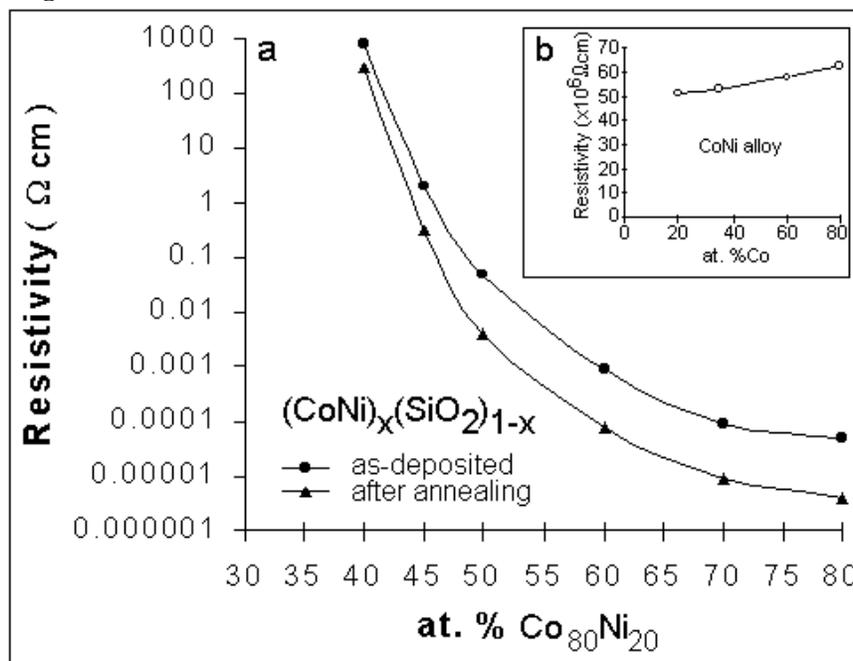


Fig. 1. (a) The CoNi content dependence of the resistivity of $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films; (b) The Co content dependence of the resistivity of CoNi alloy thin films.

The X-ray diffraction (XRD) patterns for as – deposited and annealed $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ (for x values between 0.40 and 0.60) thin films are presented in Fig. 2.

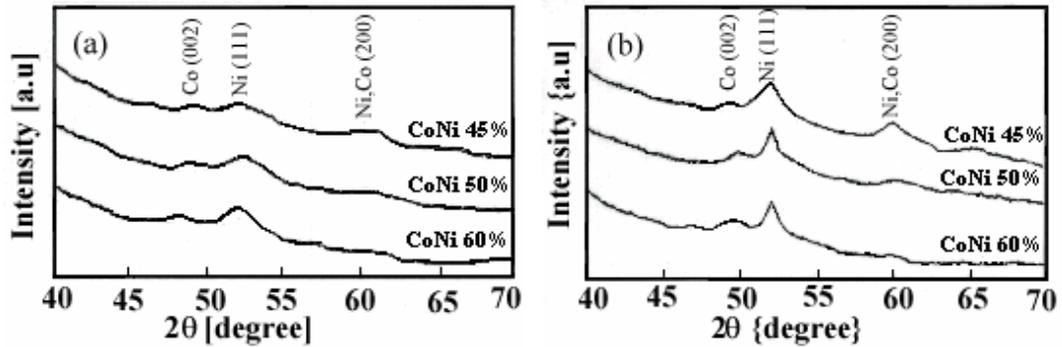


Fig. 2. X-ray diffraction patterns of the $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films: (a) as-deposited; (b) after thermal treatment at 300°C in vacuum for 1h.

The X-ray diffraction was used in order to examine the crystal structure of the CoNi particles. The XRD patterns of the as-deposited $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films samples show a nanocrystalline structure with the broadened peaks (Fig.2a). The X-ray diffraction patterns corresponding to the $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films after successively annealing in vacuum, at temperatures up to 250°C do not exhibit evident changes. After annealing at 300°C the X-ray diffraction patterns show changes in microstructure for all samples. The increasing sharpness of the diffraction peaks after annealing at 300°C is caused by the metal grain growth. Co and Ni form a continuous solid solution in the c.f.c. phase [5]. This is evidenced by the diffraction peak at $2\theta = 60$ deg., corresponding to the (200) c.f.c. Ni, Co reflections.

In Fig. 3 are presented the electron micrographs of $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ for two representative compositions: Fig. 3(a) - $(\text{Co}_{80}\text{Ni}_{20})_{0.60} - (\text{SiO}_2)_{0.40}$; Fig. 3(b) - $(\text{Co}_{80}\text{Ni}_{20})_{0.45} - (\text{SiO}_2)_{0.55}$.

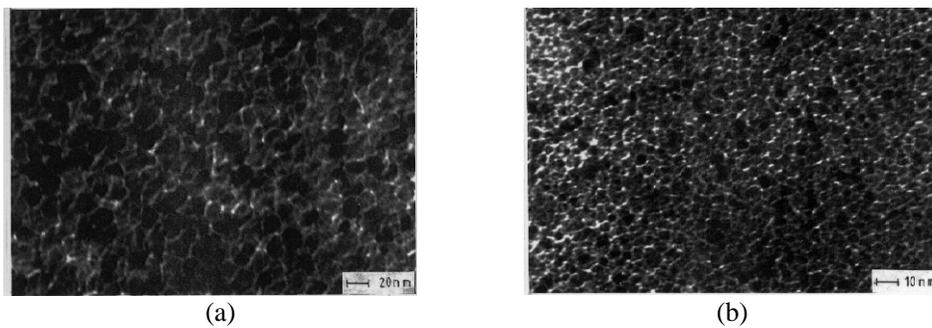


Fig. 3. Electron micrographs of $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films: (a) $(\text{Co}_{80}\text{Ni}_{20})_{0.60} - (\text{SiO}_2)_{0.40}$; (b) $(\text{Co}_{80}\text{Ni}_{20})_{0.45} - (\text{SiO}_2)_{0.55}$.

The electron micrographs of $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films (Fig.3) evidence a granular structure as follows: chains interconnected metal grains (10-20 nm in diameter) and dispersed insulator grains which fill the free spaces between the chains for $(\text{Co}_{80}\text{Ni}_{20})_{0.60} - (\text{SiO}_2)_{0.40}$ thin films; small metal grains (4-6 nm in diameter) embedded into a SiO_2 matrix for $(\text{Co}_{80}\text{Ni}_{20})_{0.45} - (\text{SiO}_2)_{0.55}$ thin films. It is clearly seen that the microstructure of $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films is strongly dependent on composition.

We measured the Hall voltages of the samples for a magnetic field applied perpendicular to the film plane. In Fig. 4 the Hall voltage dependence on the magnetic induction of the CoNi thin films is presented.

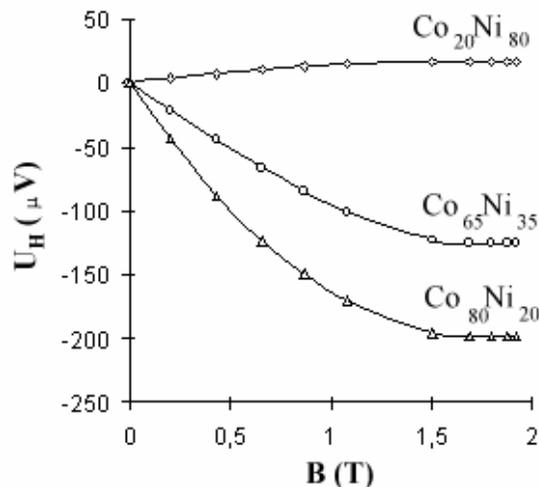


Fig. 4. The Hall voltage dependence on the magnetic induction for CoNi thin films.

The Hall voltage increases with increasing Co content in CoNi thin films. An optimum of Hall voltage values was obtained for the $\text{Co}_{80}\text{Ni}_{20}$ thin films. In Fig. 5 the Hall voltage dependence on the magnetic induction of the $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films is presented.

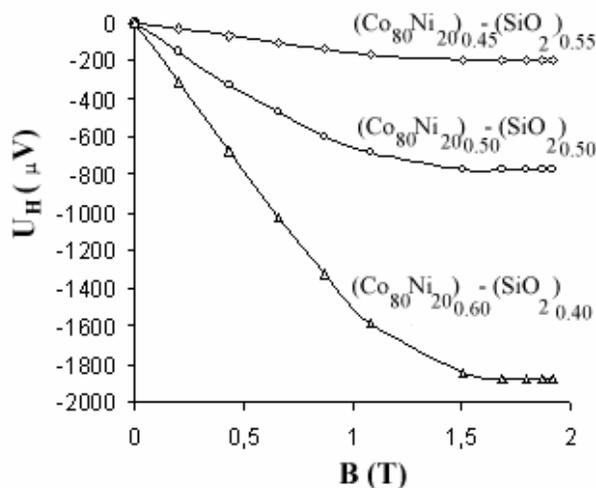


Fig. 5. The Hall voltage dependence on the magnetic induction for $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ (for x values between 0.40 and 0.60) thin films.

4. Discussion

Concerning the structure and phase evolution of as-deposited and annealed $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films the investigations described here allow the conclusion that this material is in nanocrystalline state.

The dependence of resistivity on the composition for (CoNi) alloy thin films and $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films is presented in fig.1. In Fig. 1 (b) one can see that for as – deposited CoNi thin films the value of the electrical resistivity values increase with increasing the Co content. The electrical resistivity for as – deposited CoNi thin films only slowly changed, fact confirmed in another paper [6].

In Fig. 1 (I) one can see that there is a sharp decrease of the resistivity after the annealing at temperature of 300°C , for the samples with $(\text{Co}_{80}\text{Ni}_{20})$ content ranging between 40 and 50% whilst the

samples with $(\text{Co}_{80}\text{Ni}_{20})$ content higher than 50%, e.g., above the percolation threshold, present a small decrease.

The surface resistivity values range between $0.2 \Omega/\square$ and about $28 \text{ M}\Omega/\square$, as the amount of (CoNi) in the films decreases up at about 45%.

The resistivity of the $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films decreased with annealing treatments. Two processes are involved in this behavior. The first, which occurs after annealing treatment up at 250°C , is the decreasing density of structural defects and internal stresses relaxation at $(\text{Co}_{80}\text{Ni}_{20})$ particles level. The second process, which occurs after annealing treatments at 300°C , implies grains growth, because recrystallization phenomena, fact evidenced by X-ray diffraction pattern (Fig. 2 b). These results show the strong influence of the metal grain size, on the electrical behavior of the metal-insulator films.

In Fig. 4 one can see that for as-deposited CoNi thin films, the Hall voltage values are remarkably enhanced when the Co contents in film increase. One can see also that the CoNi thin films with different compositions present an unusual behavior. The very small but positive Hall voltage value corresponding to the sample with a small Co content (about 20 at.). The CoNi thin films, with a Co content higher than 20% at. present negative values for of the Hall voltages.

In Fig.5 we present the dependence of the Hall voltage on the magnetic induction, for different compositions near the percolation threshold, for $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films. We found an important increase of the Hall voltage for as-deposited $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films with a $(\text{Co}_{80}\text{Ni}_{20})$ content of about 60%. The high difference between the magnitudes of Hall voltage for CoNi thin films and $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films can be observed, confirming the influence of the microstructure and composition on their galvanomagnetic characteristics.

5. Conclusions

The electrical and galvanomagnetic properties of the $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films show specific dependencies on the microstructure length scale. This has been demonstrated for electrical and galvanomagnetic characteristics of $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films. The electrical resistivity values range between 10^{-5} and $10^3 \Omega \text{ cm}$, as the amount of metal (CoNi) in the films decreases. An important increase of the Hall voltage with increasing $(\text{Co}_{80}\text{Ni}_{20})$ content in the $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films was observed, as compared to the (CoNi) thin films. By analyzing the presented preliminary results concerning galvanomagnetic characteristics can be seen that $(\text{Co}_{80}\text{Ni}_{20})_x - (\text{SiO}_2)_{1-x}$ thin films are interesting for magnetic sensors based on the Hall effect. The properties found in these films suggest that the galvanomagnetic properties should be optimized by further investigations of films after annealing at different temperatures.

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