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Structural and magnetic behaviour of DC sputtered Alnico type thin films

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Hard magnetic thin films of Alnico alloy having different thickness were deposited on a ceramic substrate by dc sputtering. The target was made of commercial Alnico permanent magnet having nominal composition Fe49Co23Al11Ni14Cu3. The amorphicity of as-deposited films has been checked by X-ray diffraction technique. Selected samples have been submitted to a controlled annealing to induce the amorphous-to-crystalline transformation. The influence of thermal treatments on the hysteresis properties has been studied as a function of thickness and microstructure on annealed samples.

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1. Introduction

A new challenge regarding the materials science is the discovery of new materials that can be integrated in devices such a large area of applications as in nano- or microtechnology, optoelectronic, or even biophysics. In all of these fields the demand of control of size and properties of the materials is increasing, so novel approaches are required regarding the production of thin films. Although rare-earth based alloys are now the references for the production of permanent magnets. Taking into account the difficulties of obtaining rare-earth based thin films, in what concerns their prevention of the oxidization, and knowing the costs of obtaining Fe-Co and Fe-Pt thin films, some effort have been devised to prepare different typologies of thin films based on Alnico composition.

A guiding principle, in what concerns the development of magnetically hard materials, is to obtain by alloying and heat treatment, a matrix containing finely divided particles of a second phase. These particles should be so small, less then a hundred nanometers diameter, that each grain contains only a single domain. Then the magnetization reversal occurs only by the rotation of the direction of magnetization "en bloc".

Te aim of the paper is to understand both mechanisms, structural and magnetic one in the development of Alnico type thin films starting from a bulk Alnico permanent magnet used like target in a sputtering deposition process. Until now just a few literature studies were carried on this subject: Ganzha and co. [1] reported on the production of films of Fe-Ni-Al-Co by vacuum evaporation on quartz, NaCl or ceramic substrate. They concluded that the films have inferior high-coercivity characteristics than the bulk, but also they report a difference in film composition (in terms of aluminium), which can be responsible of a different mechanism of phase transformations with respect to the bulk material. More recently, Hadjipanayis et al. [2] tried to obtain such films on silicon substrates by sputtering starting from a commercial Alnico 5 magnet. They reported higher coercivity with respect to the bulk, but – as reported by the author himself – this result is of difficult to explain, because the source of such a high coercivity can not be related with the low anisotropy of the crystalline phases formed after the annealing treatments. Therefore, in order to appreciate any development occurring in the production of Alnico thin films, it is necessary to understand the mechanisms of both magnetization processes and phase formation starting from the knowledge already developed on the corresponding bulk material.

2. Experimental

The thin films were prepared DC magnetron sputtering technique [3] using an ATC 2200 AJA International Inc. deposition system. As substrates, we used 0.5 cm^2 and 1 cm^2 of double-polished ceramic plates. The substrates were held at room temperature during deposition. In order to have a uniform elemental deposition, the target was rotated during all the experiments. The chamber base pressure was in the range of 2.2×10^{-7}mbar to 7.7×10^{-7}mbar, and the Ar plasma pressure was 5×10^{-3}mbar; the dc power was 150W. The target was made by conventional casting method starting from raw elements in order to obtain the desired thin films. The target composition was calculated to obtain a little imbalance in Fe-Co-Al elements with respect to the commercial Alnico 5 as a mean in order to optimize appropriately the composition of the produced films.

In order to evaluate the rate deposition it was used a simple conventional technique by tracing with a marker line on the substrate surface before the deposition process, then the films were immersed for 1h in acetone solution in order to remove the marker line; in this way it has been created a step measurable using an atomic force microscope instrument (see Fig. 1). Then knowing the thickness of the deposited film and the period of the deposition process it was calculated the rate deposition.

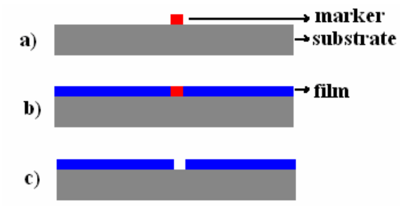


Fig. 1. A schematic representation of a) the substrate line marked before deposition process, b) the film after deposition process, c) the film after removing the marker line.

Since the deposition chamber was not opened during the experiments, all the obtained films were considered to have the same deposition rate. In order to confirm the thicknesses measured by atomic force microscopy also X-ray reflectivity (XRR) technique was used. The differences in thickness measurements considering the both techniques used were of order of 2 to 5 nm.

X-ray diffraction (XRD) and X-ray reflectivity (XRR) have been performed on the films with the Cu Kα wavelength in a parallel beam configuration. XRD patterns have been collected with an angle of incidence of the X-ray beam on the sample surface fixed at 1^0. X-ray diffractogram of the target has been performed in the Bragg-Brentano configuration.

Annealing has been done in a furnace in vacuum at low level pressure, at about 6×10^{-6}mbar, at 600 ^oC for 1h, then slow cooled to the room temperature, as presented in Fig. 2. After annealing the films were structurally investigated by XRD and scanning electron microscopy (SEM). Both, in plane and perpendicular to the plane of the films measurement were performed using vibrating sample magnetometry technique (VSM).

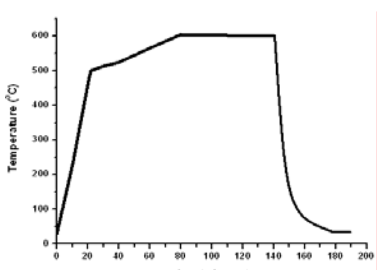


Fig. 2. Thermal treatment diagram.

3. Results and discussions

After annealing, XRD patterns reveals a crystalline structure formed mainly of cubic structures, evidenced also by SEM images (see Figs. 3 and 4). It was noticed the presence of two cubic phases Al-Fe-Ni and Fe_5Co_3, and of a deformed cubic to a tetragonal $AlNi_3$ phase. It was clearly observed that all the crystalline phases seems to be pronounced for the thickness of 150 nm of the annealed film.

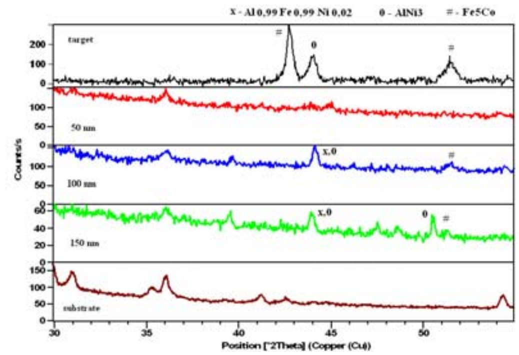
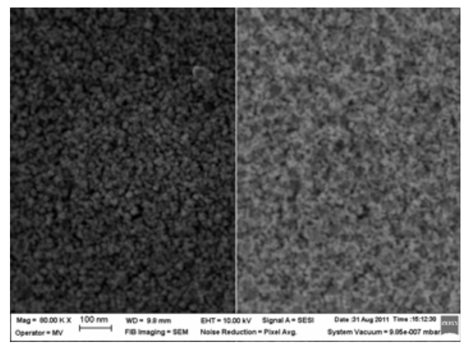


Fig. 3. XRD patterns for the target (black), the annealed films (red 50 nm thickness, blue 100 nm thickness, green 150 nm thickness) and for the ceramic substrate (brown pattern).



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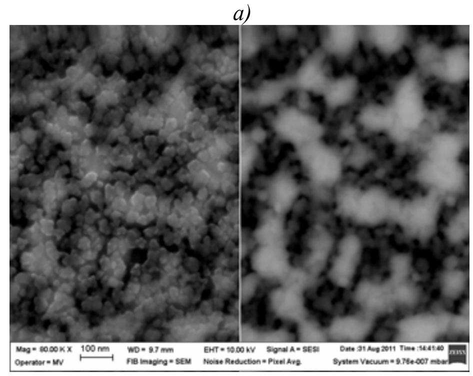

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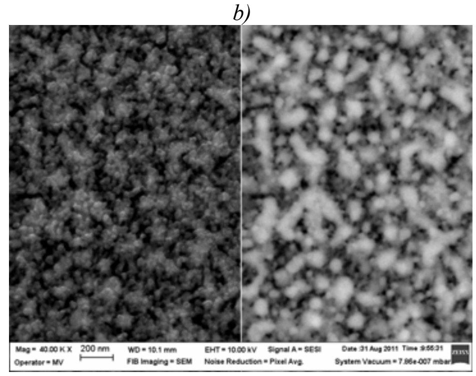


c)

Fig. 4. SEM (left secondary electron, right backscattering electron) images: a) 50 nm thickness, b) 100 nm thickness , c) 150 nm thickness.

This observation could confirm the existence of a preliminary α-phases, as previously mentioned in literature [4]. SEM images showed a compact structural arrangement of the annealed films, revealing some granular cubic grains of around 10 to 30 nm, sometimes interconnected. backscattering scanning electron (BSE) images correlated to the secondary electron images showed small differences in what concerns the elemental distribution for the involved crystalline phases. Regarding the microanalysis, it is difficult to make a clear difference between the phases formed and assess if they are richer in one or another element (in terms of Co or Ni in particular), because of the instrumental spatial resolution is comparable with the grain dimension. The presence of the formed phases could be resolved by an further TEM analysis, but this involves also choosing a convenient method to prepare the samples for TEM analysis, without changing the real structural and magnetic behaviour of the films.

Low values of coercive field, saturation induction, and consequently of maximum energy product with respect to the Alnico bulk, could be related to the incomplete phase decomposition or the structural interaction. Since during the annealing treatments an external magnetic field was not applied, the random distribution of the formed Fe-Co phase could not lead to a coercive field as high as in the bulk, where FeCo crystal growth is strongly influenced by the external field. It has to be mentioned that we would not expect that the higher value of the coercive field could be reached trough the anisotropy of the system since the phases involved have a cubic structure. The anisotropy constant of the developed magnetic structure was estimated to be of the order of 10^5 J/m^3.

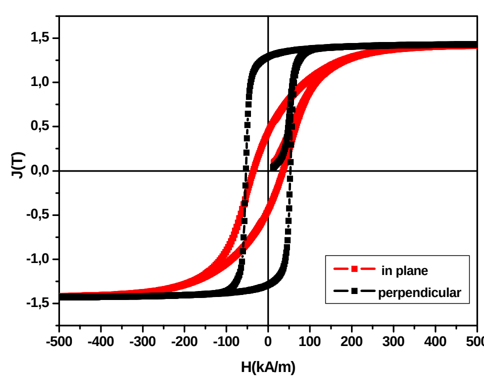
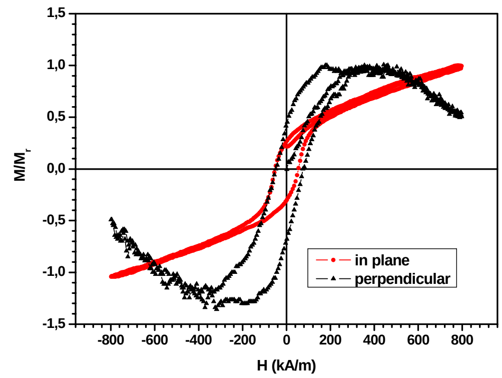


Fig. 5. Hysteresis loops of the Alnico target.

Despite the low coerciviy values for the 100 nm and 150 nm thickness films, those of 50 nm showed the highest coercivity when measured in plane and also, unexpected, out of plane. Taking into account the shape of the samples measured, thin films, usually it is expected that the in plane component of the magnetization should be stronger than the perpendicular one, however all the annealed films presented a higher coercivity perpendicular to the plane of the films. The rectangularity factor M_r/M_s lower than 0,4 indicates the possible use of this kind of films like recording media. It remains to further establish their stability, taking into account the excellent corrosion resistance and thermal stability of the bulk Alnico.



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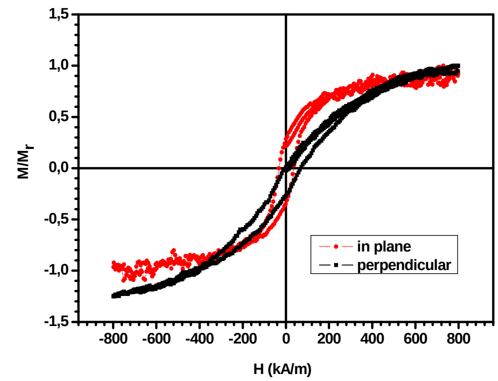

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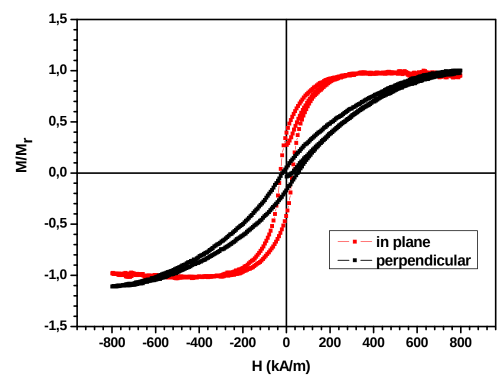

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Fig. 5. Hysteresis loops of the annealed thin films: a) 50 nm thickness, b) 100 nm thickness, c) 150 nm thickness.

Table 1. Magnetic properties of the annealed films comparatively with the Alnico target.

Thickness (nm)	Mr/Ms in plane	Hc in plane (kA/m)	Hc perp. (kA/m)
target	0.3037	36.57	53.57
50	0.3261	53.43	61.72
100	0.3366	33.90	35.79
150	0.3908	27.39	34.94

4. Conclusions

Alnico thin films can be competing candidates concerning the costs and the easy way of producing them. The structural behaviour related to the magnetic properties presented in this paper could be a good start in configuring new materials at low scale level in order to be implemented like ferromagnetic components in some micro devices. It still remains to explain the perpendicular component of the magnetization correlated to the intimae structure of the films by involving new characterization techniques and also to try to configure an ordered structure after further magnetic field assisted annealing.

Acknowledgments

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