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# Preparation and physical properties of [NdFeB/(NbCu)]× n thin films

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*Abstract*. In this paper we present our results concerning the influence of the NdFeB and NbCu layers thickness and composition on the microstructure and magnetic properties of [NdFeB/(NbCu)] × n films. By controlling the preparation and annealing conditions, an improvement in the hard magnetic properties of [NdFeB/(NbCu)] × n films can be achieved.

*Key words*: thin film; permanent magnets; spacer layer.

## 1. Introduction

Thin film NdFeB permanent magnets are important for future advances in microsensors, micromotors and microelectromechanical systems (MEMS) [1]. The miniaturisation of these microdevices requires magnetic thin films with high coercivity and remanent magnetization in order to provide strong forces [2]. Optimization of microstructure is very important for the improving of the hard magnetic properties of NdFeB thin films. Refinement of the microstructure to less than 30 nm is essential to obtain high coercivity and high-energy product [3].

A number of preparation parameters including annealing temperature, sputter gas pressure, NdFeB layer thickness, sample composition, spacer layer and underlayer materials can affect the microstructure of the NdFeB thin film and its magnetic properties [1]. Many metallic elements can be added to the ternary NdFeB system with the aim of enhancing the magnetic properties. Among them, Cu and Nb are known to act in the sense of promoting the formation of nucleation sites and suppressing the grain growth, respectively [4].

We present in this report our results concerning the influence of the NdFeB and NbCu layers thickness, layers composition, multilayer constitution and annealing parameters on the microstructure and magnetic properties of NdFeB multilayer films. A comparison between the microstructure and magnetic properties of NdFeB single layer and [NdFeB/(NbCu)]×n multilayer films is reported.

## 2. Experimental

NdFeB and NbCu thin films were prepared using a conventional R.F. diode sputtering system (Laboratory Sputtering Plant Z-400). The NdFeB sputtering target was made of a Fe plate having 7.5 cm in diameter, on which Nd and B chips were disposed. The NbCu sputtering target was made of a Nb plate having 7.5 cm in diameter, on which Cu chips were disposed. The optimal composition for the NdFeB thin films was obtained by modifying the number of chips disposed on the surface of the targets. For NbCu layer, the surface ratio was kept constant at 75/25 (%). The thickness of NdFeB layer from multilayer system was varied from 90 to 270 nm, the thickness of NbCu layer from multilayer system was fixed at 5 nm and the total thickness of NdFeB layers was of about 540 nm. The thickness of the NbCu underlayer and over layer was fixed at 20 nm.

The [NdFeB/(NbCu)] $\times$ n thin films were deposited on Si substrates mounted on a water cooled electrode and switched between different positions over the sputter guns with the NdFeB and spacer layer materials to build up multilayered films.

The structure of [NdFeB/(NbCu)] $\times$ n films was investigated by X-ray diffraction analysis using a X-ray diffractometer in a Bragg-Brentano arrangement with a monochromatized Co-K $\alpha$ radiation. For estimation of the grains size we used the Warren – Averbach method [5].

The microstructure of NdFeB thin films was investigated by transmission electron microscopy (TEM), using standard 'microscope grids' coated with an evaporated carbon thin film (with thickness of about 10 nm) as substrates. $(NbCu)_x/(NdFeB)_y/(NbCu)_x$ films with a thickness of about 70 nm (x=5 nm, y=60 nm) were used for TEM analysis.

The magnetic properties of all the samples were determined at room temperature using a vibrating sample magnetometer (VSM) in a magnetic field of up to 1600kA/m applied along and perpendicular to the film plane.

The multilayer films used for magnetic measurements and X-ray diffraction analysis are of the form $(NbCu)_x/[(NdFeB)_y/NbCu)_z]/(NbCu)_x/Si$, where x = 20 nm, y = 540, 270, 180, 90 nm and z = 5 nm.

The as – deposited [NdFeB/(NbCu)] $\times$ n films were devitrified by annealing in vacuum, for different periods of time (10÷20min.), at temperatures between 500$^o$C and 750$^o$C, to obtain optimized hard magnetic properties.

## 3. Results and Discussion

In Fig.1 are presented X-ray diffraction patterns of as-deposited NdFeB films for different compositions of sputtering target. The optimal composition for which the NdFeB thin films are amorphous was obtained for a ratio 20/67/13 (%) of surfaces occupied by the Nd, Fe and B components on the surface of the sputtering target.

Fig.2 shows the X-ray diffraction patterns for NdFeB thin films, in as – deposited state and after annealing at 650$^o$C. The as – deposited NdFeB and [NdFeB/(NbCu)] $\times$ n films, as well as after annealing at temperatures below 500$^o$C, have an amorphous structure (fig.2a). By annealing between 500$^o$C and 750$^o$C, the NdFeB and [NdFeB/(NbCu)] $\times$ n films undergo a transition from amorphous to crystalline structure and the diffraction peaks are identified as corresponding to $Nd_2Fe_{14}B$ tetragonal phase (fig.2b).

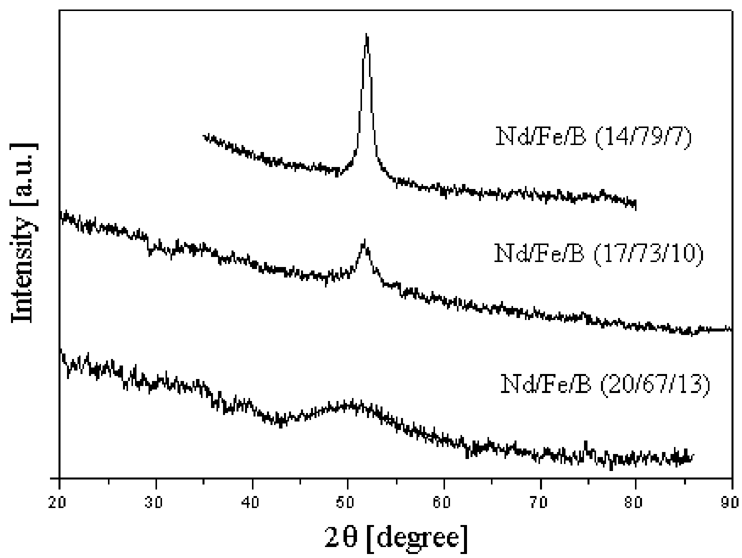


Fig. 1 - X-ray diffraction patterns of as - deposited NdFeB films for different compositions of sputtering target. The surface ratio is noted within brackets.

In the case of NdFeB single layer films annealed at 650°C for 10 min., the grains of $Nd_2Fe_{14}B$ phase are about 40 nm in diameter. For [NdFeB/(NbCu)] × n films the grains of $Nd_2Fe_{14}B$ phase reach a size of about 20 nm and some Nb- and Cu- secondary phases appear. We have concluded that a combined addition of Nb and Cu as a spacer layer with a thickness of 5nm is effective in reducing the grain size within the NdFeB layer.

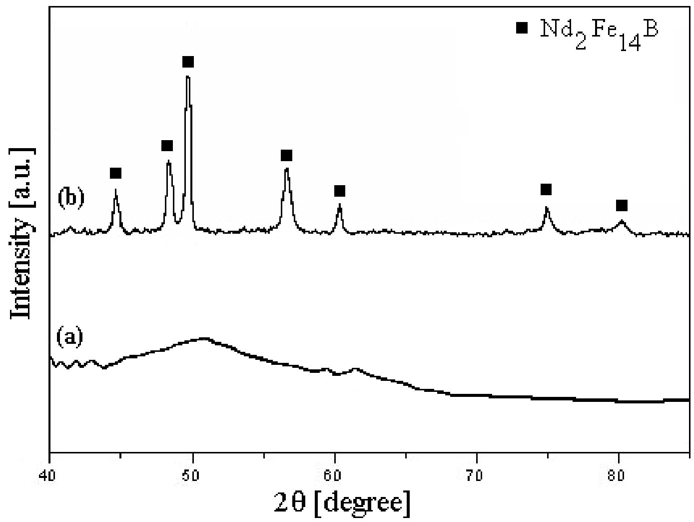


Fig. 2 - X-ray diffraction patterns of NdFeB films: (a) as-deposited and annealed at temperatures T<500°C and (b) annealed at T=650°C. The ratio in % of surfaces occupied by the Nd, Fe, B components on the surface of the sputtering target is 20/67/13.

Fig. 3 shows two electron micrographs for NdFeB (60nm) and NbCu (5nm)/NdFeB (60 nm)/ NbCu (5nm) films after annealing under vacuum for 10 min., at 650°C.

If these samples are annealed at 650°C for 20 min., or 700°C for 10 min., an increase of the grain size occurs.

The hard magnetic properties of the multilayer [NdFeB/(NbCu)] × n films are strongly influenced by NdFeB layer thickness. The effect of spacer layer on multilayer [NbCu/NdFeB(540nm)] x n films has been also investigated through the variation of magnetic properties.

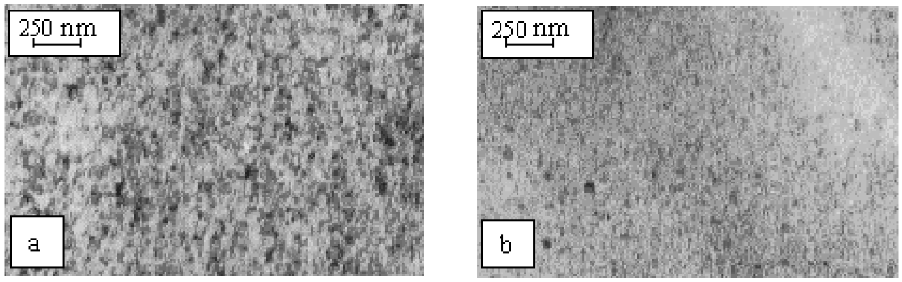


Fig. 3 - Electron micrographs for NdFeB (a) and NbCu / NdFeB (b) thin films annealed at 650ºC for 10 min.

Figs 4 and 5 present the dependence of the magnetization and coercivity, at room temperature, on the annealing temperature for NdFeB single layer and sandwiched (NbCu)/NdFeB/(NbCu) film and multilayer [NdFeB/(NbCu)] × 3 film.

In Fig. 4 one can observe that the magnetization at maxium available applied field decreases with the annealing temperature above 550°C for all samples.

All as - deposited samples present soft magnetic properties with coercivity $H_c$ < 5kA/m at room temperature for applied magnetic field in plane of the samples. For the samples annealed at about 550°C the coercive field values were found to be 20kA/m for NdFeB single and sandwiched films and about 225kA/m for multilayer [NdFeB/(NbCu)] × 3 films. At annealing temperatures between 550 and 650°C for 10 min., an increase of coercivity values for all samples has been obtained.

The multilayer [NdFeB (180 nm)/(NbCu)] × 3 system presents an enhanced coercivity as compared to the NdFeB single layer and sandwiched (NbCu)/NdFeB/(NbCu) film. The high coercivity of [NdFeB (180 nm)/(NbCu)] × 3 films was obtained due to the diffusion of Nb and Cu into multilayer system during annealing, since that these atomic species form very small grains at the 2:14:1 grain boundaries. This effect is optimum when structuring the film into multilayer form. The coercive field $H_c$ increases to 1200kA/m due to the domain wall pinning effect within the intergranular region.

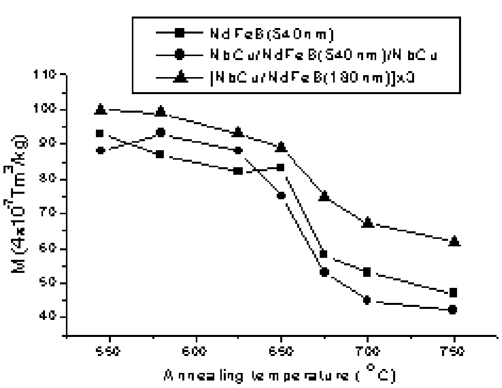

Fig. 4 - The dependence of the magnetization on the annealing temperature for 10 min., for NdFeB and multilayer [NbCu/NdFeB]x3 films

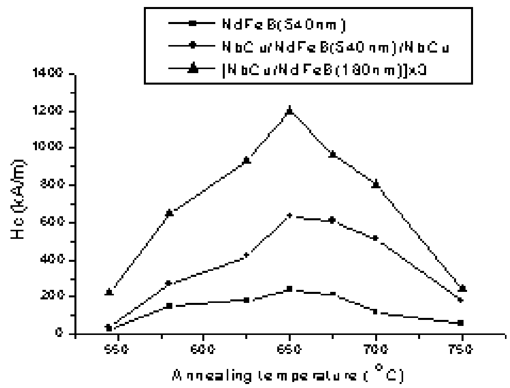


Fig. 5 - The dependence of the coercivity on the annealing temperature for 10 min., for NdFeB and multilayer [NbCu/NdFeB]x3 films

Fig. 6 presents the dependence of the remanence ratio $M_r/M(H_{max})$ on the annealing temperature for NdFeB single layer and multilayer [NdFeB/(NbCu)] $\times$ n films. $M_r$ denotes the remanent magnetization and $M(H_{max})$ denotes the value of the magnetization at the maximum applied field. Values larger than 0.5 are associated with the existence of exchange interraction between the gains [6]. The values of $M_r/M(H_{max})$ for the investigated samples range from 0.45 to 0.82. For NdFeB single layer films deposited directly on Si substrate, the remanence ratio $M_r/M(H_{max})$ values are approximately constant (=0.50) over all annealing temperatures domain.

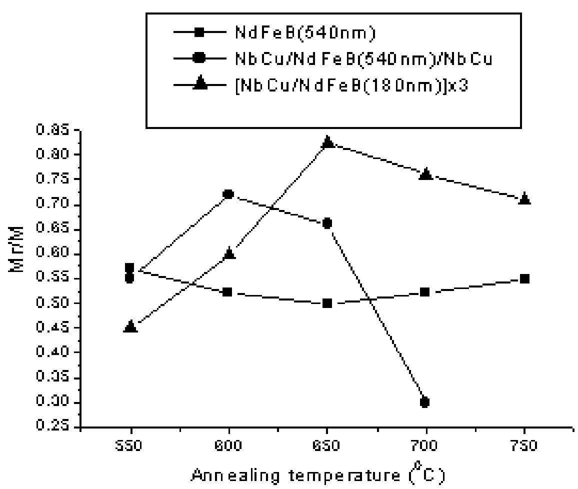

Fig. 6 - The dependence of the remanence ratio $M_r/M(H_{max})$ on the annealing temperature for NdFeB and multilayer [NbCu/NdFeB]x3 films

For NbCu/NdFeB(540nm)/NbCu and multilayer [NbCu/NdFeB(180nm)]x3 films the remanence ratio $M_r/M(H_{max})$ presents a maximum of 0.72 for an annealing temperature of $600^oC$ and a maximum of 0.82 for an annealing temperature of $650^oC$, respectively. For NbCu/ NdFeB/NbCu samples annealed at temperatures higher than $650^oC$, the remanence ratio $M_r/M(H_{max})$ decreases as a consequence of grain growth within the NdFeB layer and the existence of secondary phase at the layer interfaces [7]. In case of [NbCu/NdFeB(180nm)]x3 films, the decrease of the remanence ratio is smooth due to an uniform mixture of $Nd_2Fe_{14}B$ grains and Nb- and Cu- based secondary phases, determined by the interdiffusional process between NdFeB layers and NbCu spacer layers.

## 4. Conclusions

We have prepared NdFeB and NbCu/NdFeB thin films on Si substrates. The annealing temperature that leads to the best-achieved magnetic properties_is in the range 550-$650^oC$, for 10 min., and depends on the underlayer NbCu presence and the form of NdFeB thin films, single layer or multilayer. The $Nd_2Fe_{14}B$ grain size decreases in multilayer [NbCu/NdFeB]×n system. Samples with the largest coercivity ( about 1,200 kA/m) have $Nd_2Fe_{14}B$ grain size in the range 25 nm - 40 nm.

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