Impact of Deposition Rate on the Structural and Magnetic Properties of Sputtered Ni/Cu Multilayer Thin Films

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Abstract: The structural and corresponding magnetic properties of Ni/Cu films sputtered at low and high deposition rates were investigated as there is a limited number of related studies in this field. 5[Ni(10 nm)/Cu(30 nm)] multilayer thin films were deposited using two DC sputtering sources at low (0.02 nm/s) and high (0.10 nm/s) deposition rates of Ni layers. A face centered cubic phase was detected for both films. The surface of the film sputtered at the low deposition rate has a lot of micro-grains distributed uniformly and with sizes from 0.1 to 0.4 μm. Also, it has a vertical acicular morphology. At high deposition rate, the number of micro-grains considerably decreased, and some of their sizes increased up to 1 μm. The surface of the Ni/Cu multilayer deposited at the low rate has a relatively more grainy and rugged structure, whereas the surface of the film deposited at the high rate has a relatively larger lateral size of surface grains with a relatively fine morphology. Saturation magnetisation, $M_s$, values were 90 and 138 emu/cm$^3$ for deposition rates of 0.02 and 0.10 nm/s, respectively. Remanence, $M_r$, values were also found to be 48 and 71 emu/cm$^3$ for the low and high deposition rates, respectively. The coercivity, $H_c$, values were 46 and 65 Oe for the low and high Ni deposition rates, respectively. The changes in the film surfaces provoked the changes in the $H_c$ values. The $M_s$, $M_r$, and $H_c$ values of the 5[Ni(10 nm)/Cu(30 nm)] films can be adjusted considering the surface morphologies and film contents caused by the different Ni deposition rates.

Keywords: Magnetic Properties; Ni/Cu Multilayer Thin Films; Sputtering Technique; Surface Morphology.

1 Introduction

Broad attention continues to advance at a growing rate in nanostructured magnetic multilayers due to their favorable and unique physical properties, such as use in read heads or magnetic sensors [1]. Among the magnetic elements, Ni is one of the most common components in a magnetic film [2–4]. It has often been served together with other magnetic [5] or non-magnetic [4, 6] elements to create and investigate different material types [7, 8] grown by various production techniques, including evaporated Ni/Cu [9] and electrodeposited NiCu/Cu multilayers [10, 11]. Although a lot of techniques have been used to produce such materials [6, 8], they were extensively grown with the sputtering method because this method leads to a coating, which is dependent on the deposition parameters and, hence, the easily tunable properties [2]. For this reason, the technique has become an attractive research area. The deposition rate may be an important and effective parameter for a typical sputtering process [12, 13]. It is indicated in the study by Yi et al. [2] that different sputtering rates can cause important effects in the structural and, hence, magnetic properties. Moreover, it is stated in the study by Hedayati and Nabiyouni [14] that one of the most effective parameters that change the roughness of the thin film surface is deposition rate. To our knowledge, although there are a few studies on the production and characterisation of Ni/Cu multilayer films produced with the sputtering technique [4, 7], the effect of low and high deposition rates of magnetic layers on the properties of the Ni/Cu multilayer has not been discussed in the literature. Therefore, the aim of this study was to characterise the 5[Ni(10 nm)/Cu(30 nm)] multilayer thin films produced by the sputtering technique with two DC magnetrons.

Saturation magnetisation, $M_s$, and remanence, $M_r$, values increased with the increasing deposition rate of the Ni layers. On the film surfaces, a significant transfiguration caused by the deposition rates of the Ni layers was observed. Consequently, the changes in the film surfaces provoked
the changes in the coercivity, $H_c$, values. It is seen that the changes observed in the surface morphologies and contents of the films caused by the different deposition rates of the Ni layers can be related to the changes in the magnetic behavior of the sputtered Ni/Cu multilayer thin films.

2 Materials and Methods

Magnetic Ni/Cu multilayer thin films were deposited on commercial acetate substrates by using a sputtering system with two DC magnetrons. The commercial Ni and Cu targets were located on each magnetron separately. The targets have high purity (99.9%) and a 50.8-mm diameter. The acetate substrate was firstly cleaned with isopropyl alcohol and then with distilled water before deposition. After cleaning, it was dried at room conditions. The distance from the substrate to the target was 12.5 cm. The deposition was achieved in a pure argon atmosphere and at a pressure value of $4.5 \times 10^{-3}$ mbar. A rotary pump and a turbomolecular pump were used to perform a pressure value of $3 \times 10^{-6}$ mbar before sending argon gas to the vacuum atmosphere. During sputtering of each film, the temperature of the substrate was at room temperature. However, the variation in the substrate temperature due to the experimental process was $\pm 5$ °C. The thickness of each Ni layer was 10 nm and that of each Cu layer was 30 nm, while the total thickness of the films was 200 nm; therefore, the films can be symbolised as $[\text{Ni}(10 \text{ nm})/\text{Cu}(30 \text{ nm})]$. The thicknesses were measured by using a quartz crystal microbalance thickness monitor during the sputtering of the films. In order to investigate the effect of the different deposition rates of the magnetic layers on the structural and corresponding magnetic properties of the Ni/Cu multilayer thin films, the deposition rates of the Ni layers were chosen as 0.02 nm/s (low) and 0.10 nm/s (high). On the other hand, the Cu layers were sputtered at 0.04 nm/s. For both of the films, the Ni layer (10 nm) was firstly deposited onto the substrate, and then the Cu layer (30 nm) was sputtered onto the earlier deposited Ni layer. Thus, the first bilayer was completed, and this procedure was repeated five times to obtain a completed film structure.

The compositional analysis of the multilayers was achieved with energy-dispersive X-ray spectroscopy (EDX; AMETEK, NJ, USA). For sample excitation in the EDX technique, an electron beam with high energy was used in order to obtain an exact value throughout the cross section of the films. The crystal structure was investigated using the X-ray diffraction (XRD; PANalytical, Netherlands) technique. The analysis was done with Cu-K$_\alpha$ radiation and by scanning the Bragg angle between 40° and 70°. The surfaces of the multilayers were investigated by using a scanning electron microscope (SEM; FEI (OR, USA), Quanta 200 FEG). Also, the structural analysis was supported by atomic force microscopy (AFM; NanoMagnetic Instruments, Oxford, UK), and more detailed information was obtained about the surfaces. The magnetic hysteresis loops were detected using a vibrating sample magnetometer (VSM; ADE TECHNOLOGIES DMS-EV9, USA) at room temperature. The diameter of the circle-shaped films was 6 mm for the VSM measurements. The loops were obtained by sending the magnetic field as parallel and perpendicular to the film plane, separately, for the determination of the magnetic easy axis. The perpendicular loop was plotted only for the film sputtered at the high rate as an example.

3 Results and Discussion

Compositional analysis achieved by EDX revealed that the rate of the atomic components in the multilayers was affected by the deposition rate of the magnetic layers. The film produced considering the low (0.02 nm/s) deposition rate consists of 5.0 at.% Ni and 95.0 at.% Cu. The analysis of the film deposited at the high rate (0.10 nm/s) showed that the film consists of 70. at.% Ni and 93.0 at.% Cu.

The XRD analysis showed that the films have a face centered cubic (fcc) structure. Figure 1 illustrates the XRD patterns of the films and the substrate. In the figure, two different peaks for the (111) and (200) planes were located at Bragg angles of 43° and 50°, respectively, and the peaks of the substrate were also labeled with asterisks. The XRD patterns show that the films were preferentially crystallised at the (111) plane. The presence of peaks in the patterns shows important similarities with the study by Yi et al. [2], which investigated the sputtered Ni film with a thickness of 100 nm. In Ref. [2], the (111) and (200) planes were also detected in the same range of measurements because Ni and Cu have almost the same lattice planes. The electrodeposited Ni/Cu multilayers examined [15] were crystallised in the fcc structure with the (111) preferential orientation, and the peak of the (200) plane arose

![Figure 1: XRD patterns of the substrate and the Ni/Cu multilayers sputtered at the low (0.02 nm/s) and at the high (0.10 nm/s) deposition rates.](image-url)
along with the (111) peak as in the present study. On the
other hand, a more polycrystalline fcc structure with a
(111) texture was detected in the study by Nacereddine
et al. [9] that investigated the evaporated Ni/Cu multi-
layers with an Ni thickness of 165 nm. That is, the peaks
of (220), (311), and (222) were detected together with the
peaks of (111) and (200) in Ref. [9], although the XRD pat-
terns of the present study have only two peaks, (111) and
(200). It can be said that there is no effect of the different
deposition rates of the Ni layers on the XRD patterns of
the films. By using the XRD data, the average grain sizes
and the grain sizes of the (111) plane calculated by the
Scherrer formula [16] were found to be 30 nm. The study
by Svalov et al. [5] declared that there is also no relation
between deposition rate and the crystallite size of (111) as
in the present study. In addition, the interplanar spacing
for these planes, $d_{(111)}$, was also calculated as 0.2090 nm.
Also, the lattice parameters were calculated as 0.3619 and
0.3595 nm for the deposition rates of 0.02 and 0.10 nm/s,
respectively. It can be clearly seen that the calculated
lattice parameters agree more with the lattice parameter
of bulk Cu and Ni ($a_{Cu} = 0.36150$ nm, $a_{Ni} = 0.35238$ nm [16]).
The recent study by Svalov et al. [5] investigated the effect
of deposition rate on XRD patterns of sputtered FeNi films
and reported that all FeNi films deposited at different depo-
sition rates have similar XRD properties, as in the present
study. It is obvious that the same XRD properties obtained
from the XRD patterns are almost the same because Ni and
Cu have nearly the same crystal structure.

Figure 2a and b shows the SEM images of the films
deposited at the low (0.02 nm/s) and high (0.10 nm/s)
deposition rates, respectively. The substrate image is also
given in Figure 2c. In Figure 2a, almost uniformly dis-
tributed grains can be observed on the film surface. The
sizes of these grains were in the range of 0.1–0.4 μm. In
Figure 2b, the number of the micro-grains considerably
decreased, and their sizes increased up to 1 μm. Some
local EDX measurements done on the grains showed that
the grains have almost the same content with the base
regions of the films. Additionally, the SEM images of both
films have some stripes, probably because of the surface
of the substrate as shown in Figure 2c. It was revealed
that the deposition rates of the Ni layers have a significant
effect on the surface characteristics of the films.

The AFM images were obtained to get a more detailed
examination of the film surfaces. Figure 3a and b shows
the AFM images of the multilayers sputtered at the low
and high deposition rates, respectively. The AFM images of
the substrate are also illustrated in Figure 3c. In Figure 3a,
the surface has a vertical, acicular and recessed mode as
shown clearly in the three-dimensional image. In addition,
this acicular mode comes into prominence as micro-grains
in the two-dimensional AFM image. On the other hand,
the surface of the multilayer deposited at the high depo-
sition rate has a relatively fine structure with relatively
less roughness as shown in Figure 3b. The relatively rough
and rugged surface structure can be seen in Figure 3a
because several light dots are in the dark regions, unlike
the AFM image of the film deposited with 0.10 nm/s. The
average $R_a$ values were measured to be 6 and 47 nm for
the films deposited at the low and high deposition rates, respectively. On the other hand, Co/Cu multilayers were produced by the evaporation technique at different deposition rates and with different thicknesses of Cu layers in the study by Tsunoda et al. [17]. It was reported that the $R_m$ values were almost constant for different thicknesses of Cu layers, but increased as the deposition rate decreased. In addition, it is feasible to compare the lateral size of the grains on the surfaces from the AFM images. As can be observed in Figure 3, the lateral size of the surface grains of the film deposited at the high rate is larger than that of the film deposited at the low rate. Similarly, lateral grain size varied with different electrolyte types as revealed in the study by Ghosh et al. [15] that reported the properties of electrodeposited Ni/Cu multilayers deposited from different electrolytes. However, the surface of the substrate is in a smooth form without an acicular or grainy structure with larger micro-grains as shown in Figure 3c. Yi et al. [2] also investigated the surface of a sputter-deposited Ni film with a thickness of 100 nm and indicated that the deposition rate of the sputtered Ni probably affects the surface roughness of the film, as in the films in the present study. More rough surfaces were obtained in the present study than in the study by Yan et al. [18], which investigated the polycrystalline Ni/Cu multilayer thin films grown on an SiO$_2$ substrate with the electron beam evaporation technique. This is most probably due to the different deposition techniques used in the present study and in Ref. [18]. In the current study, even if the deposition rate of the top Cu layer was kept constant, it was assumed that the top Ni layers have a capacity to change the surface morphology. It is seen that the differences in the film surfaces are relevant to the different deposition rates, which may allow the changing energy of the Ni atoms during the sputtering process. That is, the different momentum values caused by the different deposition rates of the Ni layers lead to different surface structures during the film formation with the sputtering technique, because implantation of sputtered atoms on a substrate to get a proper film formation is relevant to the intensity of the collisions between the substrate and the sputtered atoms. Electrodeposited Ni films produced at different growth rates caused by different current densities were investigated in Ref. [14] and found that the roughness of the surface increases when the growth rate is increased, unlike in the present study.

The hysteresis loops of the films were plotted between $\pm 5$ kOe (at high field; Fig. 4a) and $\pm 500$ Oe (at low field; Fig. 4b) separately. The $M_s$ values, which were calculated considering the whole multilayer volume, were detected as 90 and 138 emu/cm$^3$ for the films deposited at the 0.02 and 0.10 nm/s Ni deposition rates, respectively.

Figure 3: Three- and two (inset)-dimensional AFM images of the multilayers deposited at (a) 0.02 nm/s and (b) 0.10 nm/s deposition rates. (c) AFM images of the acetate substrate.
The values agree with the compositional analysis of the films as the \( M_s \) value depends mainly on the type of ferromagnet and, hence, the percentage of the magnetic constituent in a film [19] besides the crystal structure [2]. In addition, it is clearly stressed in the study by Pereira et al. [20] that \( M_s \) also depends on the microstructural properties, such as grain size and lattice strain. From a different point of view, the \( M_s \) value increases with the decrease of Ni contents in electrodeposited NiFeCu alloys [20]. It should also be stated that the \( M_s \) value of bulk Ni is 480 emu/cm\(^3\) as indicated in Ref. [19]. According to this value, the calculated \( M_s \) values are higher than the expected values when Ni contents were considered. This deviation from linear dependence between the \( M_s \) values and Ni contents may arise from the surface grains detected in the SEM images. A similar effect arising from grain boundary segregation of the crystallites was also reported in our earlier study [21]. Besides, a typical film structure may cause and support the relatively high \( M_s \) values with respect to that of the bulk Ni. The \( M_s \) values were also detected from the hysteresis loops. The \( M_s \) values were found to be 48 and 71 emu/cm\(^3\) for the films sputtered at the low and high deposition rates of the Ni layers, respectively. Although \( M_s \) is not directly dependent on \( M_s \) [19], the increase in the \( M_s \) value was attributed to the increase in the \( M_s \) value because the values are proportional with the \( M_s \) values. The \( H_c \) values were found to be 46 and 65 Oe for the low and high deposition rates, respectively. The \( H_c \) values are higher than those of the electrodeposited NiFeCu alloys obtained in the study by Pereira et al. [20] but lower than those of the electrodeposited Ni-Cu/Cu multilayers investigated in the study by Hedayati [22]. It can be stated that the multilayer became magnetically slightly harder when the deposition rate of the Ni layers increased. The change in the \( H_c \) values can be attributed to the change on the film surface homogeneities indicated in the SEM and AFM images because the XRD data analysis is almost the same for the films. It is clearly indicated in Ref. [20] that \( H_c \) can be affected by changes in the structural properties and film content, besides the structure of the grain boundary [2]. It should also be remembered that the change in the \( H_c \) values depends on the surface homogeneities of the magnetic materials [19]. Namely, the more micrograins caused by the vertical acicular structure lead to relatively softer magnetic properties as compared to the other surface structure. Also, a higher \( H_c \) value may be prevented by a relatively low Cu content in the Ni/Cu film deposited at the high rate because Cu is a non-magnetic element [19]. Similarly, the \( H_c \) values gradually decreased as the Cu content decreased in study by Hedayati [22] that investigated Ni-Cu/Cu multilayers electrodeposited with different thicknesses. Also, the reported \( H_c \) values in Ref. [22] are higher than those of the present study. The study by Ergeneman et al. [23] also indicated that there is a considerable relation between surface morphology and magnetical hardness of a magnetic film. Also, it was shown in the study by Gągorowska et al. [7] that the \( H_c \) and \( M_s \) values of the Cu/Ni multilayers increase gradually and almost with the same dependence as the Ni layer thickness increases. As shown in Figure 4, the perpendicular hysteresis loop of the film deposited at the high rate has a lower \( M_s \) value (24 emu/cm\(^3\)) with a higher \( H_c \).
value (285 Oe) and also a relatively gradual increase in magnetisation than that of the in-plane loop. This can be attributed to the magnetic shape anisotropy and displays that the easy-axis direction of the magnetisation is in the film plane. The same response of magnetisation to the direction of the external field was reported for the Ni/Cu films investigated in Ref. [9] and for the electrodeposited NiCu/Cu multilayers studied in Refs. [10, 11].

4 Conclusion

The effect of different deposition rates on the correlation of surface morphologies–magnetic properties of the $5\text{[Ni(10 nm)/Cu(30 nm)]}$ multilayer thin films was investigated for the low (0.02 nm/s) and high (0.10 nm/s) deposition rates of Ni layers. To our knowledge, this effect has not been widely investigated earlier for sputtered Ni/Cu multilayers, and there are a few related studies on this topic in the literature. In this study, a good platform has been provided for the presentation and comparison of the obtained results. The films crystallised in the fcc phase. On the film surfaces, a significant transfiguration caused by the different deposition rates of the Ni layers was observed in both the vertical and lateral sizes of the surface grains. The $M_s$ and $M_r$ values changed with the change of film contents caused by the change of the deposition rate of the Ni layers. The more grainy and rugged structure caused by the vertical acicular morphology of the film sputtered at the low deposition rate of the Ni layers leads to the relatively softer magnetic properties. In addition, the changes in the film surfaces provoked the changes in the $H_c$ values. The changes in the surface morphologies and contents of the films caused by the different deposition rates of the Ni layers lead to changes in the magnetic behavior of the sputtered Ni/Cu multilayer thin films.

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