International Journal of Physical Sciences Vol. 7(46), pp. 6044-6052, 9 December, 2012
Available online at http://www.academicjournals.org/IJPS
DOI: 10.5897/IJPS12.480
ISSN 1992 - 1950 ©2012 Academic Journals

Full Length Research Paper

Composition dependence of structural, morphological and magnetic properties of Co (FCC)-Cu granular films

P. Sirisangsawang[1], W. Rattanasakulthong[1]* and S. Pinitsoontorn[2]

[1]Department of Physics, Faculty of Science, Kasetsart University, Bangkok, 10900, Thailand.
[2]Department of Physics, Faculty of Science, Khon Kean University, Khon Kean, 40002, Thailand.

Accepted 23 November, 2012

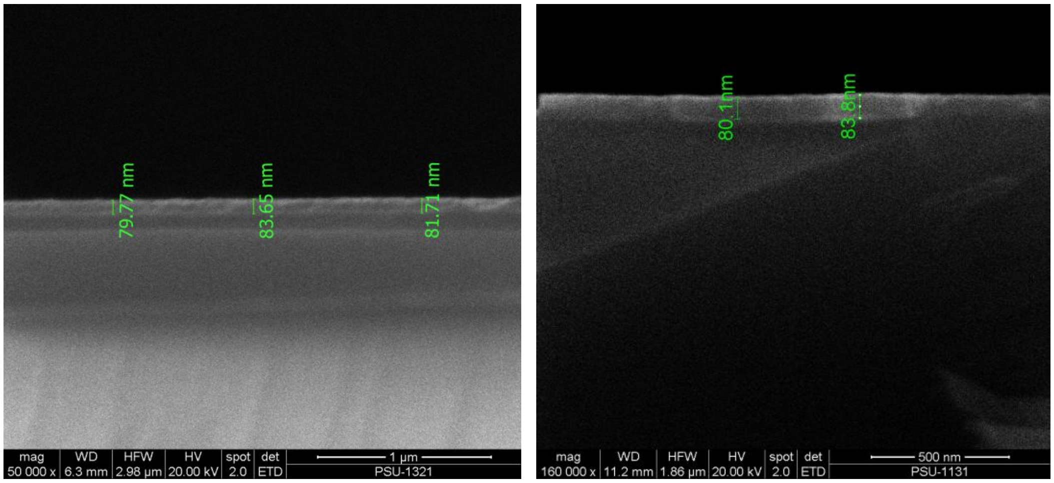
The Cobalt (Co) (FCC)-Copper (Cu) magnetic films with different compositions (x = 55.5, 44.4, 22.5) grown on glass substrate are prepared by sputtering process using a Co target and Cu sheets. The effects of the composition on morphological, structural and magnetic of the films were reported. Scanning electron microscopy (SEM) results showed that the film thickness is about 80 nm. Atomic force microscopy (AFM) measurements revealed that surface roughness of Co-Cu films is in the range of 1 to 6 nm. From vibrating sample magnetometer (VSM) result, it clearly shows that not only remanent and saturation magnetizations are increased with Co fraction but also coercivity and squareness. All results revealed that the increase of Co fraction gives rise to the lower surface roughness and the higher coercive field because of the Co coalescence and their distribution in Cu matrix.

Key words: Sputtered Co-Cu films, granular films, roughness, hysteresis.

INTRODUCTION

Magnetic granular films are composed of the ferromagnetic granules [Cobalt (Co), Iron (Fe) and Nickel (Ni)] embedded in non-magnetic metal matrixes [Copper (Cu), Silver (Ag) and Gold (Au)]. These films have attracted much interest because of their applications in giant magneto-resistive (GMR) sensors and high density magnetic recording media. The granular films have intensively been developed for perpendicular magnetic recording media in hard disk drives (HDDs) technology. These films contain small magnetic particles with a high coercive field and perpendicular magnetic anisotropy. Granular films can be synthesized by using various techniques; for example, radio frequency (RF)-sputtering (Sugawara et al., 1998; Andres et al., 1999; Tuan et al., 2003; Hiep et al., 2007), arc melting (Majumdar et al., 2009), electro-deposition (Pattanaik et al., 2003; Safak et al., 2006), and ion beam deposition (Khan, 1997; Errahmani et al., 2001; Islam et al., 2008). Because of GMR effect and numerous applications from immiscible alloying elements, sputtered Co-Cu granular films have widely been studied by many research groups such as Andres et al., 1999; Tuan et al., 2003; Errahmani et al., 2001; Hiep et al., 2007). Various kinds of underlayers substrates, substrate temperatures and high temperature annealing have been tested in order to reach the high coercivity and perpendicular anisotropy for magnetic recording applications. Errahmani et al. (2001) reported that the substrate temperature showed an opposite effect compare to the heat treatment. Moreover, GMR and saturation field was decreased with increasing substrate temperature and Co-Cu granular films synthesized by an ion-beam sputtering technique (IBS) with a high substrate temperature which have a significant mobility and quasi-segregation with large grain size. For Co-Cu films sputter-deposited on Si (100) substrates, the coercive field was significantly dependent on the annealing temperature and the perpendicular anisotropy was found in Co-rich films (Tuan et al., 2003). Additionally, the thickness consistently plays an important role on magnetic properties of magnetic thin films. Thickness-dependent coercivity and magnetization of the magnetic films on various substrates have been studied (Haque et al., 2001; Islam et al., 2008; Sharma et al., 2009). Haque et al. (2001) found that thermal evaporated Ni films on n-GaAs substrate exhibited an anomalous magnetization

*Corresponding author. E-mail: fsciwrr@ku.ac.th. Tel: +66 25 625555. Fax: +66 29 428029.



(a) **(b)**

Figure 1. SEM micrographs of (a) Co and (b) sputtered $Co_{54.5}Cu_{45.5}$ granular films.

with temperature in parallel field because of an interaction at interface of Ni and GaAs substrate, but Ni films on Si substrate did not display remarkable anomaly. Co and Fe films with the thickness of 10 to 200 nm prepared by e-beam evaporation technique on GaAs (001), coercivity of Co and Fe films was decreased with an increase in the film thickness up to threshold values of 50 and 40 nm, respectively. After threshold thickness, coercivity was almost linearly increased with an increase of the thickness up to 125 and 100 nm for Co and Fe films, respectively. Beyond 125 and 150 nm of Co and Fe films, respectively, coercivity did not change with an increase of thickness (Islam et al., 2008). In addition to a threshold thickness, they remarked that coercivity of Fe films was enhanced because of surface roughness, while the enhanced coercivity of Co films was due to a lateral interaction between Co films and GaAs substrate. Sharma et al. (2009) also reported that Co films with the thickness of 5 to 100 nm on GaAs(100) substrate showed amorphous phase at a lower thickness and then became a crystalline at higher thickness. Lately, RF-sputtered Co films on X-ray lithographic micro-pillars (Sukonrat et al., 2011) and on glass substrate (Chanthong et al., 2012) display the dissimilar magnetic properties. Co films on patterned substrate show a soft ferromagnetic phase with a higher saturation magnetization, whereas the Co films on glass substrate have a harder ferromagnetic phase with higher coercive field.

In this work, structural, magnetic and morphological properties of sputtered Co-Cu granular films on glass substrate prepared by RF-sputtering using Co target with Cu sheets were investigated to reveal the effect of Co fraction on the surface roughness, structural and magnetic properties. Hysteresis loops were performed in which current is in-plane and perpendicular to magnetic field in order to verify magnetic anisotropy of the Co-Cu granular films.

MATERIALS AND METHODS

Co-Cu granular films with different compositions were deposited on unheated glass substrates by RF-sputtering in Ar atmosphere. The composite target prepared from Co (99.99%) target and Cu (99.95%) sheets was installed at a distance of ~4.5 cm away from the substrate. The base pressure in the chamber was around 10^{-5} mbar and the argon pressure during the deposition was 10^{-3} mbar. Before coating process, the target was cleaned by glow discharge process. Variation of the composition of sputtered Co-Cu films was controlled by an involved number of Cu pieces placed on the Co target. $Co_{54.5}Cu_{45.5}$, $Co_{44.3}Cu_{55.7}$, $Co_{22.5}Cu_{77.5}$, $Co_{14}Cu_{86}$, $Co_{9.8}Cu_{90.2}$, $Co_{6}Cu_{94}$ and $Co_{1.7}Cu_{98.3}$ granular films were prepared from Cu sheet for 1, 2, 3, 4, 5, 6, and 7 pieces placed on a Co target, respectively. The deposition process was performed with constant sputtering power of 200 W at sputtering time for 1 h. The chemical composition of sputtered Co-Cu films was identified by an energy-dispersive (EDS) detector using line-scan profiles. Structural, magnetic and morphology properties of the sputtered films were studied by X-ray diffraction (XRD) with CuK_{α} radiation, vibrating sample magnetometer (VSM) under an applied magnetic field of 10 kG and atomic force microscopy (AFM), respectively. Scanning electron microscopy (SEM) was used to investigate the thickness of Co-Cu granular films.

RESULTS AND DISCUSSION

Figure 1 shows examples of side view SEM micrographs of Co and Co-Cu films and smooth deposited interfaces. The thickness of sputtered Co-Cu film grown on the glass substrate from SEM measurement is estimated as 80 nm. Examples of EDS analysis of (a) $Co_{54.5}Cu_{45.5}$ and (b) $Co_{1.7}Cu_{98.3}$ films are shown in Figure 2. The EDS result also indicates an existence of an oxide content or amorphous phase in a higher Co fraction films. This result is corresponding to amorphous phase of Co films

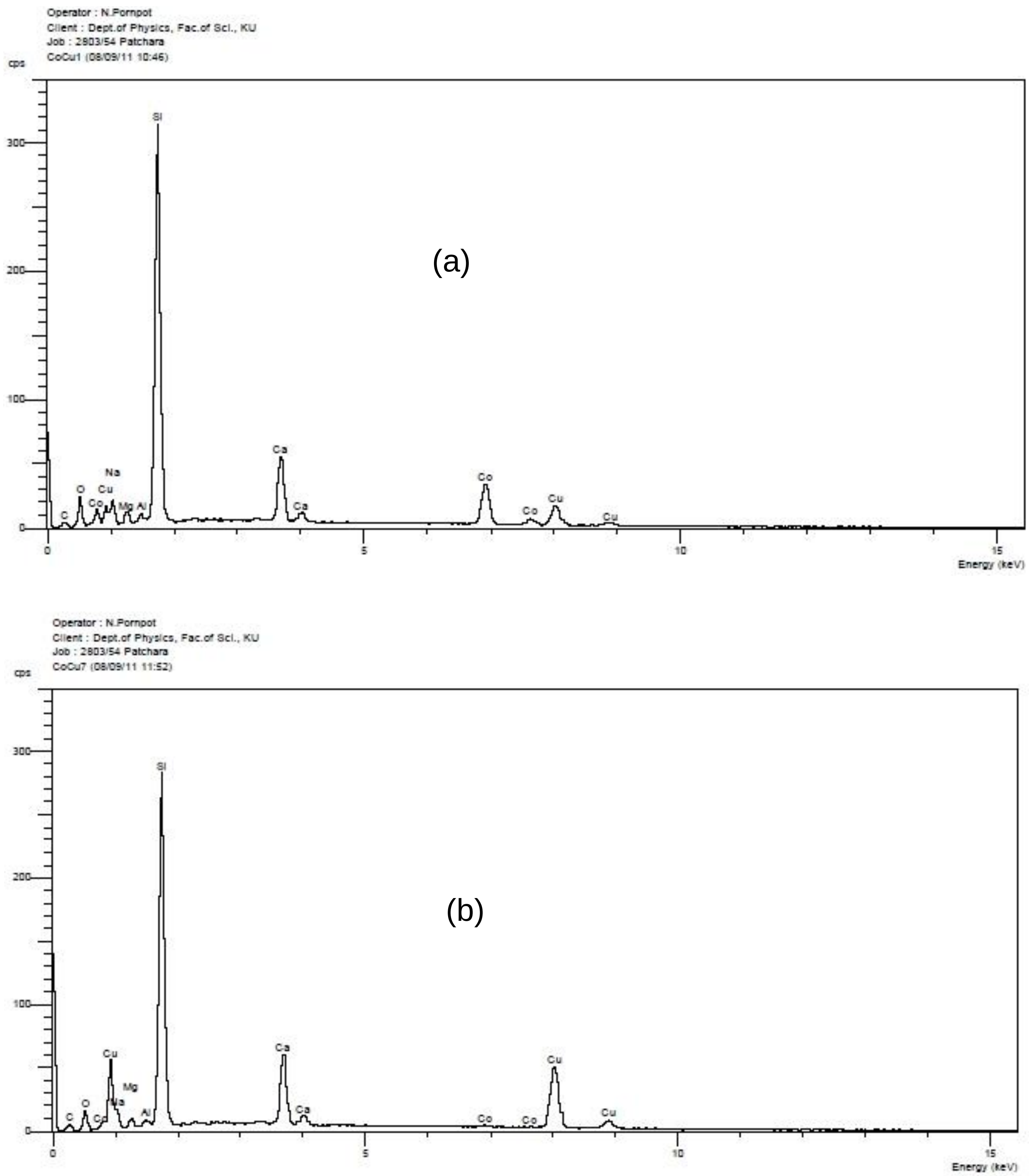


Figure 2. Examples of EDS results of (a) $Co_{54.5}Cu_{45.5}$ and (b) $Co_{1.7}Cu_{98.3}$ films.

at a low thickness reported by Sharma et al. (2009). XRD patterns of Co film and Co-Cu films are displayed in Figure 3. A FCC-Co peak with (111) orientation is observed in sputtered Co film. As-sputtered Co-Cu films show two major peaks of FCC- Cu (111) and FCC-Co (111) phases in the case of Co fraction >22.5%. For Cu-rich films (Co fraction <22.5%), FCC-Cu (111) peak and the formation of copper oxide peak are observed, whereas the FCC-Co (111) phase disappears because of the existence of Co-amorphous phase. These XRD peaks imply that sputtered Co-Cu films are consisted of Co granules homogeneously imbedded in Cu matrix for Co fraction >22.5%. The intensity of Cu peak is obviously increased with increasing Cu fraction. Amorphous phase

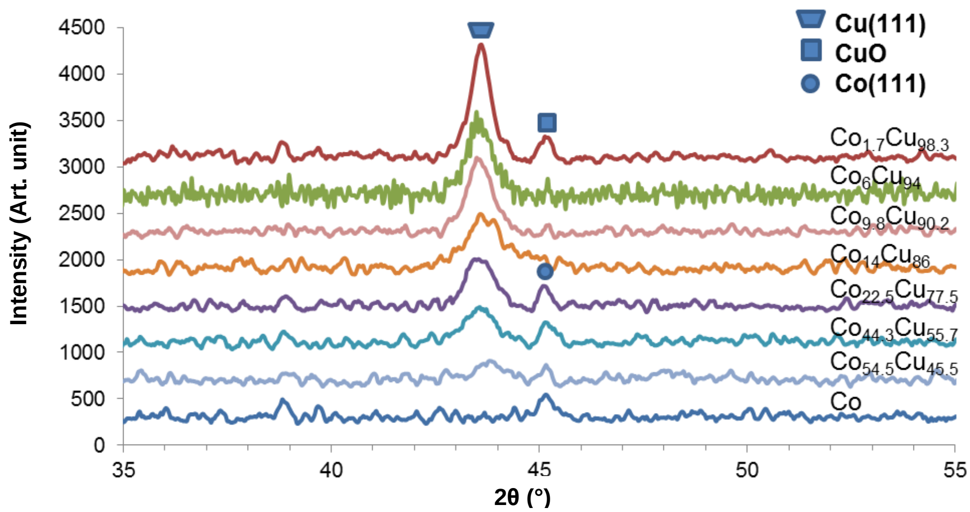


Figure 3. X-ray diffraction patterns of sputtered Co and Co-Cu films.

of Co granules are slightly increased with increasing Co fraction giving rise to an almost constant intensity of Co peak. This result agrees with composition analysis by EDS. It can be described that the oxide phase is slightly increased with increasing Cu fraction. With increasing of Co fraction, the Cu peak is slightly shifted to higher 2θ angle. It signifies that Cu lattice parameter is reduced and the strain effect occurs during the dispersion of fine Co clusters into Cu matrix. From these results, it can be concluded that Co-Cu granular films with various compositions can be prepared by using the Co target and Cu sheets.

Figure 4 shows the AFM images of sputtered Co-Cu films with different compositions over scan areas of 1 × 1 μm. The surface morphology reveals granular structure with different grain size, distribution and shape with varying of Co fraction. A tendency of cluster formation is evidently observed in Figure 4a and b. It clearly shows that these films are composed of islands of Cu and Co clusters. The smoothest surface is found in $Co_{54.5}Cu_{45.5}$ film with surface roughness (R_a) of 0.85 nm and the highest surface roughness of 6.20 nm is observed in $Co_{9.8}Cu_{90.2}$ film. The Co grain size embedded in Cu matrix seems to be bigger in higher Co-fraction films and the Co film is mainly composed of the biggest islands with continuous grains on its surface, whereas the $Co_{14}Cu_{86}$ film consists of a regular spherical shape with an almost uniform distribution. From these AFM results, it can be confirmed that grain size, shape and distribution are dependent on the composition. Average surface roughness (R_a) of Co-Cu granular films with various Co fractions is plotted in Figure 5. Because of a higher sputtering rate of Cu atoms than that of Co atoms, the variation of surface roughness with Co fraction is probably due to the difference of accumulative rate between Co atoms and Cu atoms during sputtering process (Yoshitake et al., 2004).

For Co-Cu films with Co fraction <10%, the surface roughness is tightly enhanced with increasing Co fraction and then reaches the maximum value at Co fraction of 9.8%. It can be described that Co atoms are started to diffuse into Cu matrix and agglomerate into the large islands in $Co_{9.8}Cu_{90.2}$ film. It confirms that Co-Cu granular film was started to form after the threshold of Co-fraction at around 9.8%. This result agrees with the XRD result that Co peaks are not observed in Co-Cu films with Co fraction < 10%. After the threshold of Co fraction (Co fraction >10%), the surface roughness is decreased with increasing Co fraction and then approaches the minimum surface roughness at Co fraction of 54.5%. The lowest roughness of $Co_{54.5}Cu_{45.5}$ film is originated from an equivalence of a conglomeration on glass substrate of Cu and Co atoms owing homogeneous surface morphology; it can be concluded from AFM results that surface morphology of Co-Cu granular films is implicitly composition dependent.

Room temperature hysteresis loops of Co and Co-Cu films with various compositions when an applied magnetic field is parallel and perpendicular to the film plane are shown in Figures 6 and 7, respectively. The ferromagnetic phase is observed in the Co-Cu films consisted of Co content >22.5%. For Co content <22.5%, the Co-Cu films exhibit the diamagnetic phase. It can be inferred that this phase is due to an amorphous content of Co phase. Magnetic parameters from hysteresis loops of Co-Cu granular films are summarized in Table 1. Both in-plane and perpendicular magnetizations reveal that not only the magnetization but also coercive field are increased with increasing Co content. The results confirm the intrinsic property of saturation magnetization. It is reliable that the saturation and remanent magnetization are obviously dependent on Co fraction in the films, while

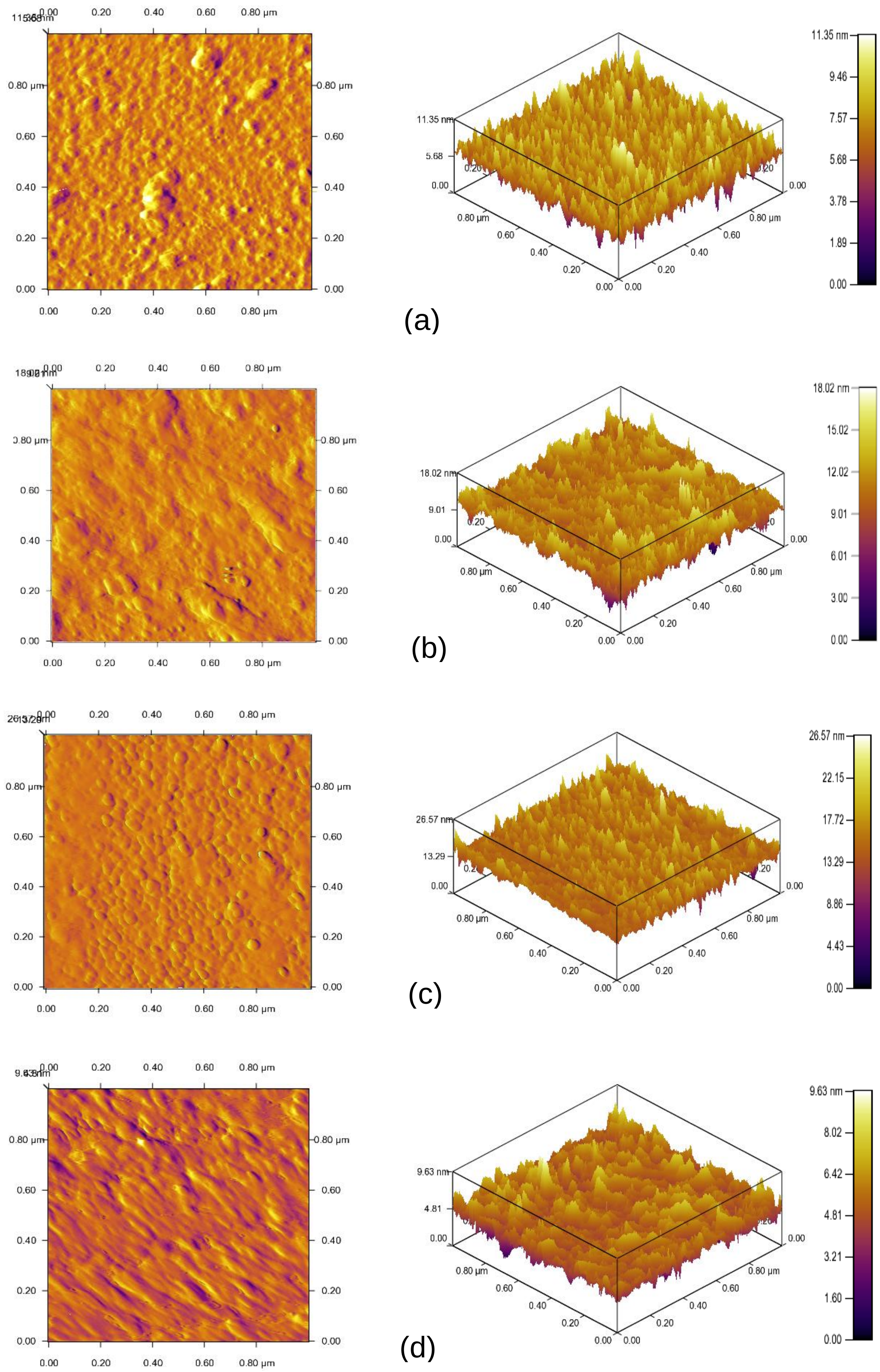


Figure 4. AFM images of (a) $Co_{1.7}Cu_{98.3}$, (b) $Co_{9.8}Cu_{90.2}$, (c) $Co_{14}Cu_{86}$, (d) $Co_{22.5}Cu_{77.5}$, (e) $Co_{44.3}Cu_{55.7}$ and (f) $Co_{54.5}Cu_{45.5}$ films.

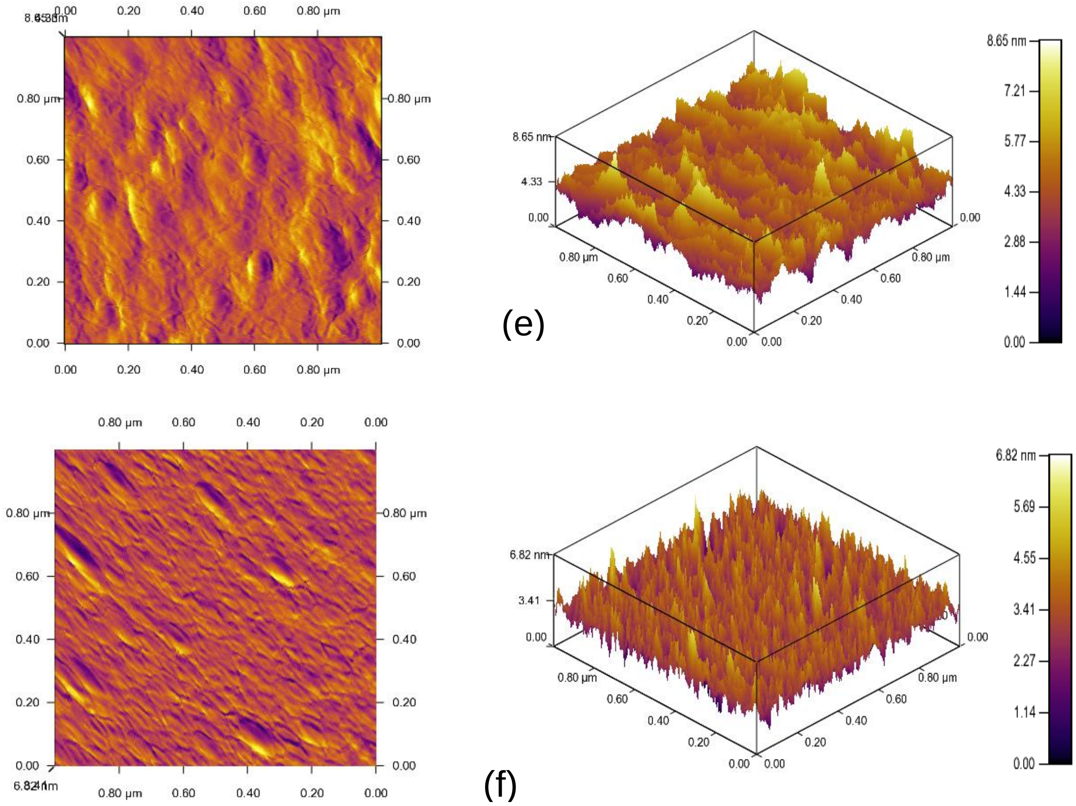


Figure 4. Contd.

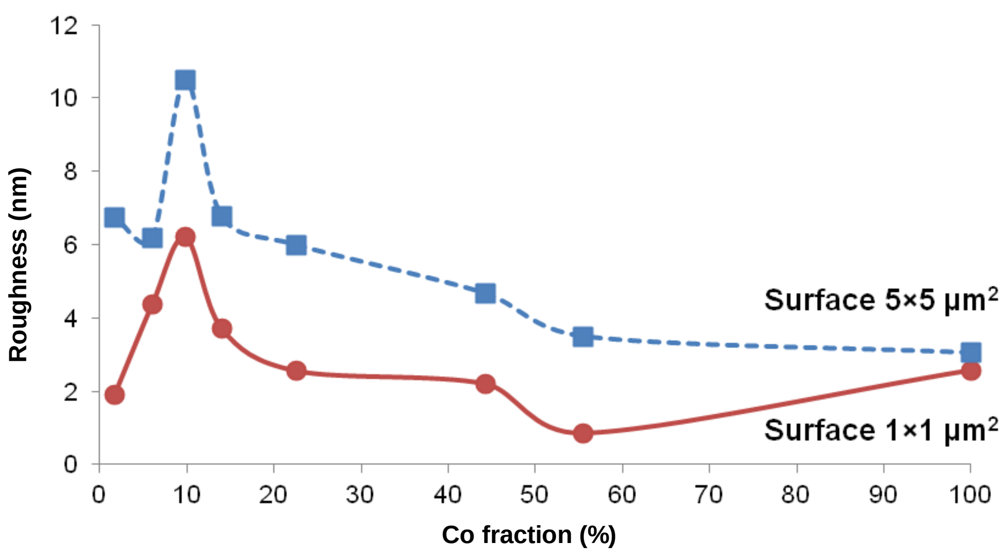


Figure 5. Surface roughness of sputtered Co-Cu films with various Co fractions in comparison to sputtered Co film.

the coercive field is strongly dependent on the magnetic grain size and distribution. Moreover, the enhanced coercivity is significantly correlated to the surface roughness because of the film composition. The enhancement of squareness is originated from the speedy increase of the saturation magnetization with

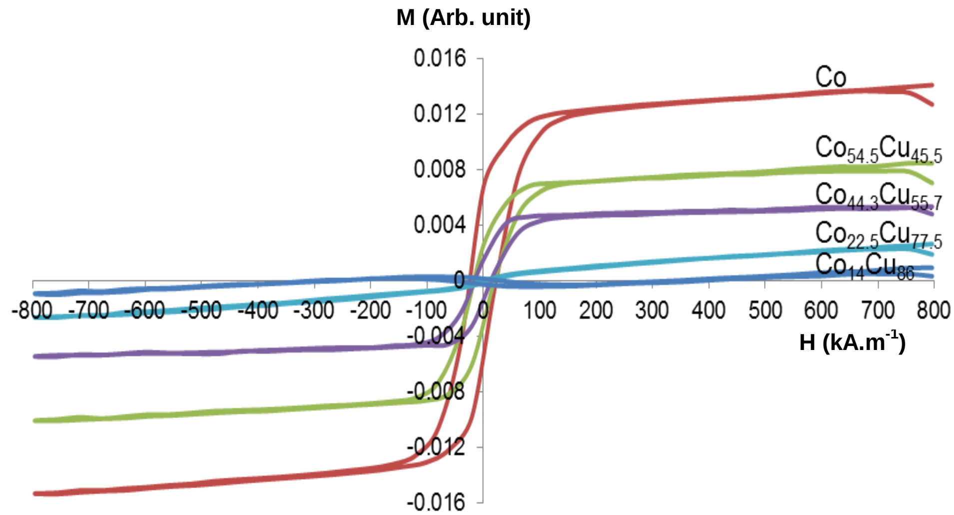


Figure 6. Hysteresis loops of sputtered Co-Cu films with different compositions when magnetic field is applied in the parallel direction to the film plane.

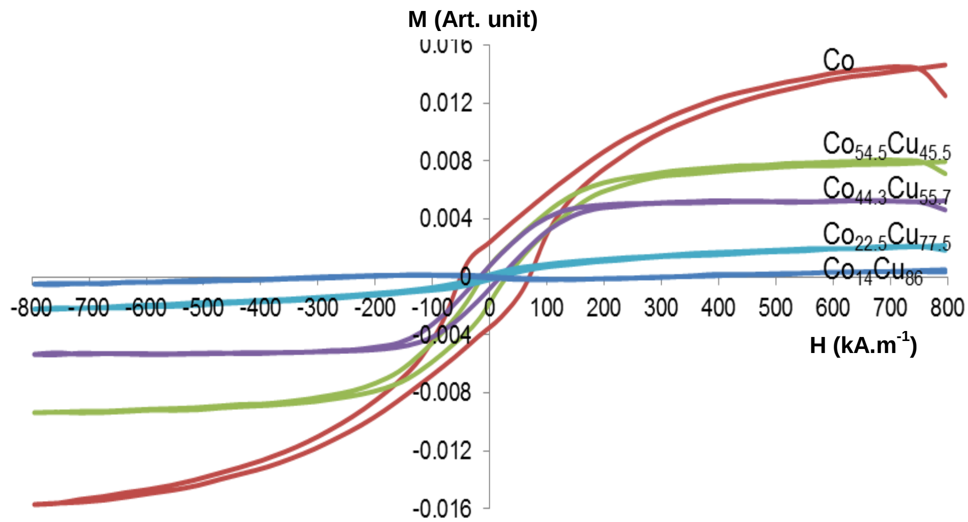


Figure 7. Hysteresis loops of sputtered Co-Cu films with different compositions when magnetic field is applied in the perpendicular direction to the film plane.

Table 1. Coercive field (H_c), saturation magnetization (M_s), remanent magnetization (M_r) and squareness (S) of Co-Cu granular films with different compositions.

Sample	Perpendicular				In-plane			
	H_c (kA.m^{-1})	M_s (Arb. unit)	M_r (Arb. unit)	$S_\perp$	H_c (kA.m^{-1})	M_s (Arb. unit)	M_r (Arb. unit)	$S_{\|}$
Co	59.91	0.0146	0.0029	0.20	23.23	0.0139	0.0061	0.44
$Co_{54.5}Cu_{45.5}$	21.30	0.0080	0.0013	0.16	17.95	0.0082	0.0028	0.34
$Co_{44.3}Cu_{55.7}$	18.31	0.0053	0.0008	0.15	14.45	0.0053	0.0015	0.27
$Co_{22.5}Cu_{77.5}$	15.33	0.0021	0.0002	0.08	4.58	0.0025	0.0001	0.02

increasing of Co fraction. The dependence of the coercive field on Co fraction is shown in Figure 8. The increase in coercive field with increasing Co content implies that Co atoms form small clusters or small particle

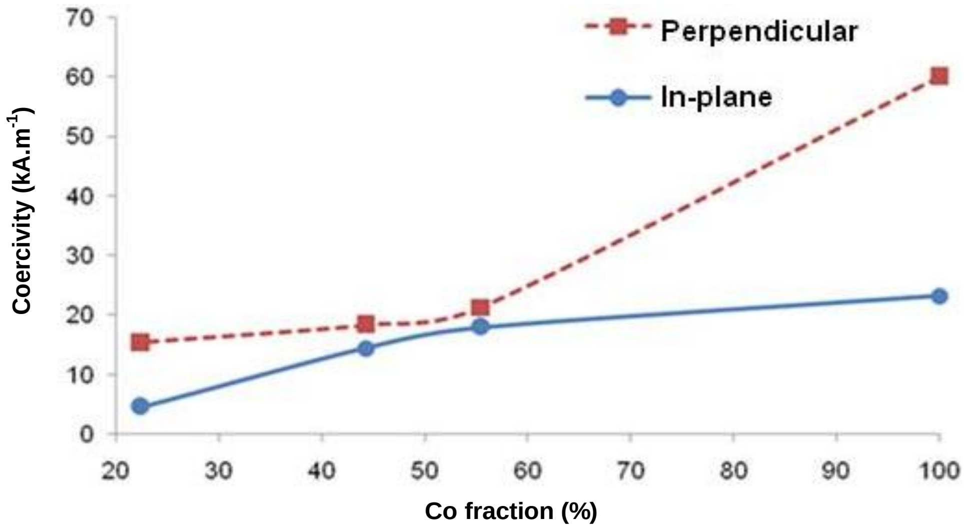


Figure 8. Coercive field of sputtered Co-Cu films with different compositions is plotted as a function of Co fraction.

in the case of low Co content. These particles then start to agglomerate into larger clusters when the Co fraction is increased as shown by AFM and the softest magnetic phase is clearly observed in $Co_{22.5}Cu_{77.5}$ films. It can be defined that Co content at 22.5% is an initial stage of the homogeneous dispersion of the small grain sizes of Co granules in a Cu matrix. From the hysteresis loops, a maximum in-plane squarness (S) of Co-Cu and Co films is about 0.33 and 0.44, respectively. The result indicates an in-plane magnetic anisotropy of the Co-Cu and Co films. The shape dependence of hysteresis loop on the composition signifies that the magnetic anisotropy of the Co-Cu granular films can be induced by the variation of the film composition.

Conclusions

The homogeneous Co-Cu films with FCC-Co (111) phase can be prepared by sputtering technique using Co target and Cu sheets for Co fraction >22.5%. The morphology and surface roughness of Co-Cu films are found strongly depended on Co fraction. Structural and magnetic properties of the Co-Cu films are considerably dependent on Co fraction or composition in films. The magnetic anisotropy of Co-Cu films tends to develop perpendicular anisotropy with increasing Co fraction. The increase of Co fraction gives rise to the lower surface roughness and the higher coercive field because of the Co accumulation and their distribution in Cu matrix.

ACKNOWLEGEMENTS

This research project was financially supported by the Industry/University Cooperative Research Center (I/UCRC) in HDD Component, the Faculty of Engineering, Khon Kaen University and National Electronics and Computer Technology Center, National Science and Technology Development Agency. The matching fund by Faculty of Science, Kasetsart University is also acknowledged.

REFERENCES

Andres JP, Colino J, Riveiro JM (1999). Enhancement of GMR in as-deposited Co-Cu granular films with RF sputtering power. J. Magn. Magn. Mater. 196-197(1-3):493-494.
Chanthong T, Rattanasakulthong W, Sirisathitkul C, Pinitsoontorn S (2012). Morphology and properties of radio frequency (RF) sputtered cobalt thin films. Int. J. Phys. Sci. 7(11):1820-1827.
Errahmani H, Berrada A, Schmerber G, Dinia A (2001). Comparative study between the effect of annealing and substrate temperature on the magnetic and transport properties of $Co_{20}Cu_{80}$ granular alloys. Mater. Lett. 51(1):48-55.
Haque SA, Matsuo A, Seino Y, Yamamoto Y, Yamada S, Hori H (2001). Anomalous temperature-dependent magnetization of Ni films on semiconductor substrates. J. Magn. Magn. Mater. 226-230(2):1591-1593.
Hiep VV, Chau N, Hong DM, Loung NH (2007). High coercivity and magnetoresistance of CoAg, CoCu granular films. J. Magn. Magn. Mater. 310(2):2524-2526.
Islam J, Yamamoto Y, Shikoh E, Fujiwara A, Hori H (2008). A comparative study of Co and Fe thin films deposited on GaAs(001) substrate. J. Magn. Magn. Mater. 320(3-4):571-574.
Khan HR (1997). Structural and magnetic properties of ion beam deposited nanocrystralline granular Co-Cu and Co-Cu-O films. J. Magn. Magn. Mater. 165(1-3):297-300.
Majumdar S, Singha RK, Yoon J, Jung MH, Chakraborty M, Das AK Das, Ray SK (2009). Microstructure and magnetic properties of melt-spun $Cu_{0.95}Co_{0.05}$ granular alloy. Phys. B. 404(12-13):1858-1861.
Pattanaik GR, Pandya DK, Kashyap SC (2003). Effect of process parameters on GMR in electrodeposited Co-Cu nanogranular thin films. Thin. Solid Films. 433(1-2):247-251.
Safak M, Alper M, Kockar H (2006). Parameters affecting microstructure

and magnetoresistance of electrodeposited Co–Cu alloy films. J. Magn. Magn. Mater. 304(2):e784-e786.

Sharma A, Trippathi S, Lakshmi N, Sachdev P, Shripathi T (2009). Thickness dependent magnetic and structural properties of Co films grown on GaAs (100). Solid State Commun. 149(25-26):1033-1038.

Sugawara T, Takanashi K, Fujimori H (1998). Appearance of GMR on annealing in Co-Cu granular alloys with high Co concentration. J. Magn. Magn. Mater. 177-181(part 2):951-952.

Sukonrat P, Sriphung C, Rattanasakulthong W, Sirisathitkul C (2011). Characterization of cobalt films on X-ray lithographic micropillars. Adv. Mater. Res. 335-336:1000-1003.

Tuan NA, Luong NH, Chau N, Hiep VV, Ha NM (2003). High coercivity and perpendicular anisotropy in Co-Cu granular films. Phys. B. 327(2-4):400-403.

Yoshitake M, Yamauchi Y, Bose C (2003). Sputtering rate measurements of some transition metal silicides and comparison with those of the elements. Surf. Interf. Anal. 36(8):801-804.