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A STUDY ON THE SOFT MAGNETIC PROPERTIES OF Fe-Ta-(N,C) NANOCRYSTALLINE THIN FILMS

Dong-Hoon Shin, *Dong-Hoon Ahn, Hyoung-June Kim, Seung-Eui Nam

Dep't of Metallurgy & Material Science, Hong-Ik University
72-1,Sangsu-Dong, Mapo-Gu, Seoul 121-791, Korea
*Dep't of Image & Media Lab, LG electronics
16, Woomyeon-Dong, Seocho-Gu, Seoul, 137-140, Korea

ABSTRACT: Magnetic properties of FeTaN and FeTaC films deposited by DC magnetron reactive sputter were investigated, and correlated with their microstructures. The optimum magnetic properties of Hc : 0.25 Oe, Bs : 14.5 kG, and μ' : 4000 (5MHz) are observed in the $Fe_{78.8}Ta_{8.5}N_{12.7}$ film, and Hc : 0.25 Oe, Bs : 14.5 kG, and μ' : 2700 (5MHz) in the $Fe_{75.6}Ta_{8.1}C_{16.3}$ film. In both FeTaN and FeTaC films with minimum grain size show the best soft magnetic properties. Thermal stability of the soft magnetic properties of FeTaN is found to be higher than FeTaC for similar compositons. TaN and TaC particles form to retard the growth of α-Fe grains. TaN particles in FeTaN films show higher efficiency in retarding the grain growth during heat treatments resulting the higher thermal stability, compared to TaC particles in FeTaC films.

I. INTRODUCTIONS

MIG type magnetic recording heads require the soft magnetic films with high magnetic flux density. Recently, Fe-based nanocrystalline films have been reported to have high saturation magnetization as well as good soft magnetic properties[1~3]. Nanocrystalline microstructures can be realized by adding N or C with nitrides or carbides forming transition metal elements(e.g., Zr, Nb, Ta, Hf). These alloying elements forms fine nitrides or carbides particles in the Fe matrix, suppressing the grain growth of α-Fe grains during the post-depostion heat treatments[4,5].

Generally, the films with C addition are known to have higher efficiency in retarding α-Fe grain growth, thus leading to better magnetic properties and higher thermal stabilities, compared to the films with N addition. However, comprehensive studies on the difference in the film microstructures and magnetic properties between FeTaN and FeTaC films have not been reported. In this study, we investigated the microstructural evolutions and magnetic properties of FeTaN and FeTaC films, which are known to have good magnetic properties, to make a direct comparison between two films.

II. EXPERIMENTS

FeTa(C or N) films with 1 μm thickness were deposited by DC magnetron reactive sputtering apparatus. Ta compositions were changed by changing the area of Ta chips on the 4" Fe target. N or C contents in the films were controlled by partial pressure of N_2 or CH_4 gases. The initial pressure was 1×10^{-6} Torr, and the working pressure was fixed at 1.5 mTorr. The N_2 or CH_4 partial pressure(P_r) was changed at 0~0.3 mTorrs(P_r/P_r+P_{Ar}=0~20%). The DC power was 420V, 0.47~0.48A and deposition rates were 500~600 Å/min, which slightly depended on reactive gas partial pressure.

Glass (Corning #7059) was used as a substrate, which was water-cooled during deposition. Deposited samples were vacuum-annealed (5×10^{-5} Torr) for 30 mins from 400℃ to 700℃. The film compositions were analyzed by Auger electron spectroscopy calibrated with a standard sample. The formation of phases and grain sizes in the films after heat treatments was investigated by X-ray diffraction and TEM studies. The coercivity (Hc) and saturation magnetization (4πMs) were measured by B-H loop tracer and VSM(Vibrating Sample Magnetometer), and the effective permeability was measured using the

Figure-8 coil method.

III.RESULTS AND DISCUSSION

Fig. 1. shows the 4πMs and Hc values of FeTaN and FeTaC films annealed at 500℃ as a function of the reactive gas partial pressure. For FeTaN films, 4πMs decreases from

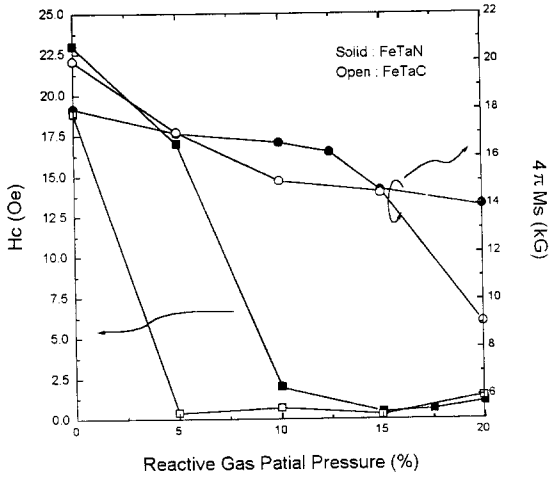


Fig. 1. Change of Hc and 4π Ms as a function of the reactive gas partial pressure.

18 kG to14 kG with increasing P_{N_2}. Hc decreases with P_{N_2} to the minimum value 0.25 Oe at P_{N_2} of 15~17%, then slightly increases at higher P_{N_2}. For FeTaC films, similarly to FeTaN films, 4πMs decreases with an increase of P_{CH_4}. High 4πMs above 14.5 kG can be obtained at P_{CH_4} up to 12.5%. The Hc more rapidly decreases with P_{CH_4} than FeTaN films and reaches the minimum values of 0.25~0.5 Oe at the P_{CH_4} of 5~15%. The process range showing minimum Hc appears to be larger in FeTaC than in FeTaN.

The behavior of Hc can be explained in terms of α-Fe grain sizes in annealed films. The average grain sizes, determined by FWHM with α-Fe (110) peaks of XRD, are plotted as a function of reactive gas partial pressure in Fig. 2. For FeTaN films, the average grain sizes slowly decrease with partial pressure and reach minimum grain sizes about 50 Å at 15%, at which films show the minimum Hc. On the other hand, in FeTaC films, grain sizes rapidly decrease to the mimimum value at 5%, without significant changes for further increase in CH_4 partial pressure. This result indicates that C addition is more effective in refining grain sizes than N addition.

The effective permeability (μ') of 5 MHz for FeTaN and FeTaC films annealed at 500℃ is plotted against various reactive gas partial pressures in Fig. 3. The behavior of change of μ' is found to be closely related

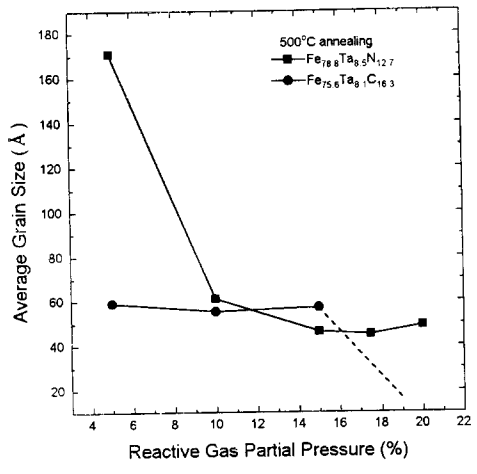


Fig. 2. Change of average grain size as a function of the reactive gas partial pressure.

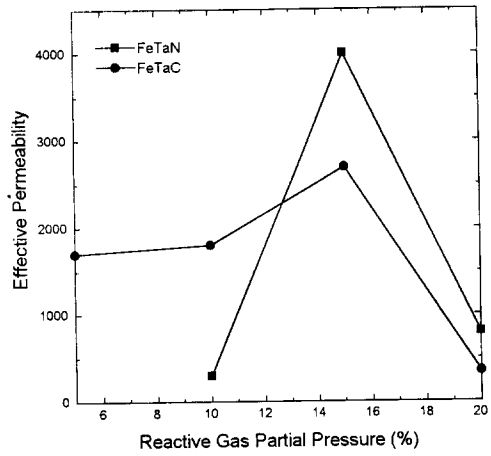


Fig. 3. Change of effective permeability as a function of the reactive gas partial pressure.

with the Hc value in both films, in that the films with low Hc show high μ'. The μ' increases with partial pressure and shows the maximum value at 15% in both FeTaN and FeTaC films. The maximum value of μ' is 4000 in FeTaN films, which is higher than that of FeTaC films(2700). The best soft magnetic properties (FeTaN ; 4πMs : 15 kG, Hc : 0.25 Oc, and μ' : 4000, FeTaC ; 4πMs : 14 kG, Hc : 0.25 Oc, and μ' : 2700)are obtained at partial pressure of 15% in both FeTaN and FeTaC films, and corresponding film compositions are $Fe_{78.8}Ta_{8.5}N_{12.7}$ and $Fe_{75.6}Ta_{8.1}C_{16.3}$

respectively.

Dependence of μ' on frequency was investigated for the films with best soft magnetic properties as shown in Fig. 4. While $Fe_{75.6}Ta_{8.1}C_{16.3}$ film shows slightly lower

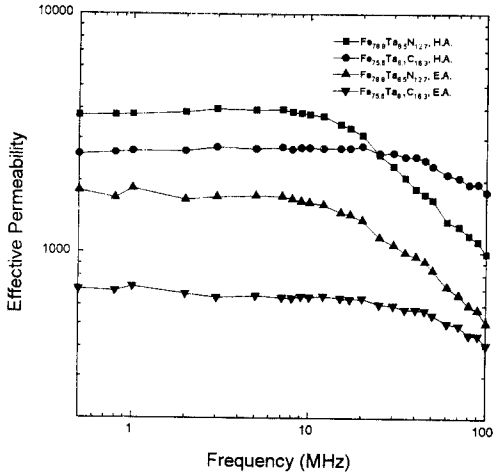


Fig. 4. Dependency of effective permeability on frequency for FeTaN and FeTaC films.

μ' value and better high frequency chracteristics, compared with $Fe_{78.8}Ta_{8.5}N_{12.7}$ film. The μ' of $Fe_{75.6}Ta_{8.1}C_{16.3}$ film sustains up to the frequency as high as 60 MHz.

The changes of magnetic properties of FeTaN(orC) films for various annealing temperatures are shown in Fig. 5.

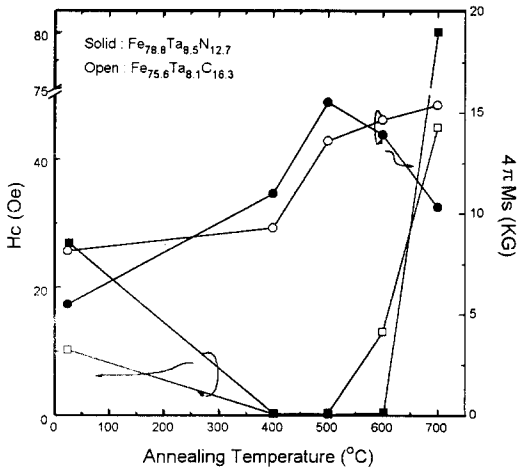


Fig. 5. Change of Hc and 4π Ms as a function of the annealing temperature.

While FeTaN films show the maximum 4πMs at 500℃, FeTaC films show a continuous increase of 4πMs up to 700℃. The Hc of films is lowered to the minimum value at 400℃ in both films. As can be seen, rapid degradation of Hc is observed above 500℃ and 600 ℃ for FeTaC and FeTaN, respectively. Similar results are observed in the investigation of μ' as shown in Fig. 6. Higher thermal stability of μ' is observed in the FeTaN films.

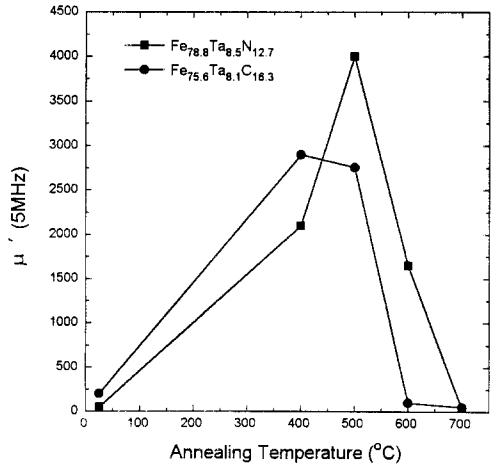


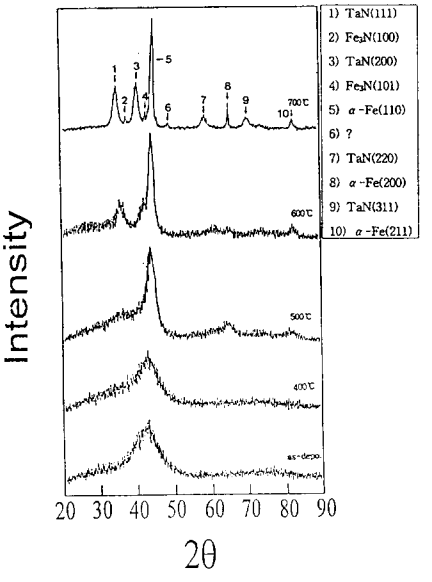
Fig. 6. Change of effective permeability as a function of the annealing temperature.

The superior thermal stability of FeTaN is somewhat in contrast with the reports that the C-added films have higher thermal stabilities than N-added films. Microstructural changes of annealed films were investigated by XRD, and the results are shown in Fig. 7. As shown in Fig. 7.(a)., as-deposited FeTaN films have amorphous structures, which hold up to 400℃. Crystallization occurs at 500℃, showing the strong α-Fe (110) textures. The broadening of the α-Fe (110) peaks could result from the particle size effects (Scherrer effects) and non-uniform expansion of BCC lattice of α-Fe with supersaturated N atoms. Asymmetrical peak shape with preferential broadening toward low diffraction angle (or higher interplanar distance) indicates that the lattice expansion. It may be the major cause of the apparent peak broadening. At 600℃, Fe_3N peaks are shown near α-Fe (110) peaks. The Fe_4N or Fe_2N phases, which are known to be the stable phases in the Fe-N phase diagrams, are not observed here[6]. The formation of Fe_3N may be related to the transient phase transformation of the supersaturated α-Fe phases. At 700℃, sharply defined α-Fe, TaN, Fe_3N peaks are observed indicating the growth of these phases.

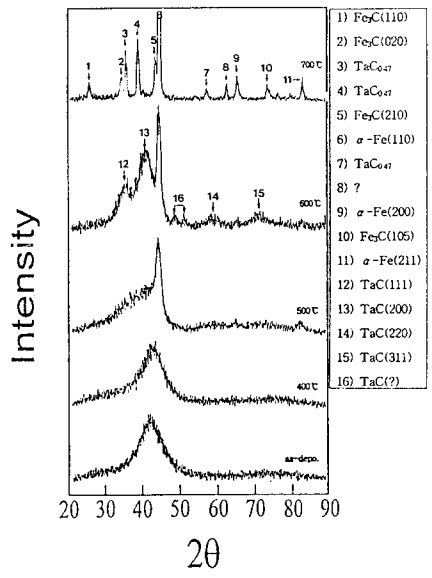
Similar behavior of microstructural changes is observed in FeTaC films, as shown in Fig. 7.(b). Apparent cyrstallization of amorphous FeTaC films occurs at 500℃,

Journal of Korean Magnetics Society, Volume 5, Number 5, October 1995

showing the strong α-Fe (110) peaks. Compared with FeTaN films, two distinct features in the diffraction peak



(a) $Fe_{78.8}Ta_{8.5}N_{12.7}$ films.

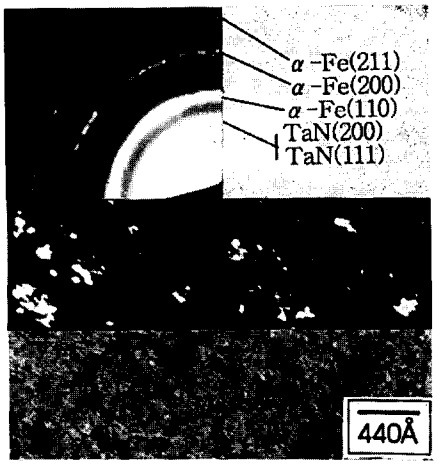


(b) $Fe_{75.6}Ta_{8.1}C_{16.3}$ films.

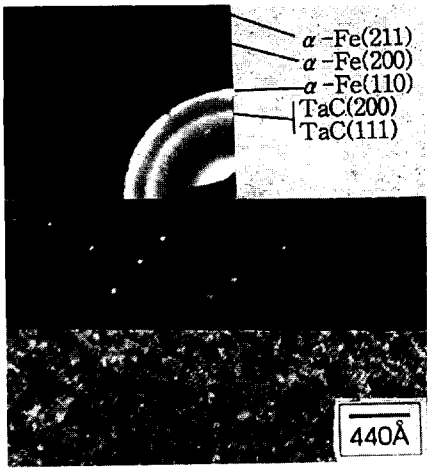
Fig. 7. XRD diffraction patterns of FeTaN and FeTaC films annealed at various temperatures.

profiles can be recognized here. Firstly, broadening of α-Fe (110) peaks are relatively lower and peak profile is more symmetrical than in FeTaN films. These peak profiles suggest that the peak brodening mainly results from the particle size effect rather than the lattice expansion. Another feature is the strong hallow pattern which exists around 30~43° diffraction angles. The hallow pattern is considered to come from the presence of fine TaC particles. At 700℃, shaply defined α-Fe, $TaC_{0.47}$, Fe_3C peaks are observed. It is interesting to note that the TaC phases, which observed at 500℃, are transformed to $TaC_{0.47}$ phases at this temperature. Detail mechanism for this transformation is not uncertain.

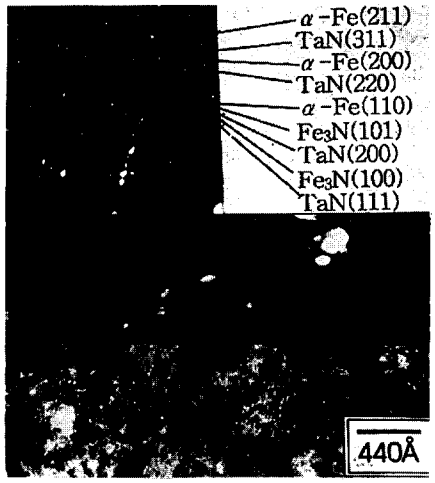
More detail information on the microstructure of



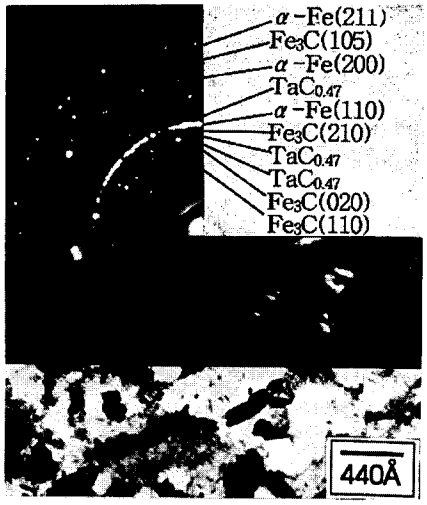
(a) $Fe_{78.8}Ta_{8.5}N_{12.7}$ films, annealed at 500℃.



(b) $Fe_{75.6}Ta_{8.1}C_{16.3}$ films, annealed at 500℃.



(c) $Fe_{78.8}Ta_{8.5}N_{12.7}$ films, annealed at 700℃.



(d) $Fe_{75.6}Ta_{8.1}C_{16.3}$ films, annealed at 700℃.

Photo. 1. TEM images and electron diffraction patterns of FeTaN and FeTaC films annealed at 500℃ and 700℃.

films can be obtained by TEM studies. As-deposited films show amorphous structures in both FeTaN and FeTaC films. Fig. 8. is the TEM micrographs with SAD patterns for the microstructures FeTaN and FeTaC films annealed at 500℃ and 700℃. At 500℃, both films show very fine α-Fe grains with sizes of 60~70 Å, which are in agreement with XRD results. The distiguishable features of SAD patterns between FeTaC and FeTaN films annealed at 500℃ are that TaC diffraction shows well defined ring pattern for FeTaC films, while TaN diffraction shows broadened ring pattern in FeTaN films. These results suggest that some coarsening of TaC particles already occurs at this temperatue in FeTaC films. Coarsening of the TaC particle will reduce the efficiency in impinging the grain growth of α-Fe. The efficiency in impinging grain growth of TaN and TaC particles can be recognized in the films annealed at 700℃. The grain sizes of FeTaC films appear to be substantially larger than those of FeTaN films. It is noted that fine grains lead to good soft magnetic properties with low Hc and high m' values, the observed higher thermal stability of FeTaN films can be explained by higher efficiency of TaN particles in suppressing grain growth.

IV. CONCLUSIONS

The magnetic properties and microstructures of FeTa-N,C films were investigated, and the results can be summarized as follows.

1) The magnetic properties of FeTaN films with Hc : 0.25 Oe, 4πMs : 14.5 kG, and μ' : 4000 (5MHz) and FeTaC films with Hc : 0.25 Oe, 4πMs : 14.5 kG, and μ' : 2700 (5MHz) can be obtained.
2) The process range showing the good magnetic properties is larger in FeTaC films than in FeTaN films.
3) In both FeTaN and FeTaC films, the films show the best soft magnetic properties at showing the minimum grain sizes.
4) TaN particles in FeTaN films show higher efficiency in retarding the α-Fe grain growth during heat treatments than TaC particles in FeTaC films, resulting the higher thermal stability of FeTaN films.

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