Microstructural Investigations of Fine Grain Co-Hf-C Films

S. Suwabe, Bunsen Y. Wong and David E. Laughlin

Department of Materials Science and Engineering,
Carnegie Mellon University, Pittsburgh, PA 15213-3890 U.S.A.

Abstract- **Microstructural investigations have been performed on Co-Hf-C films, which show high μ(>2000), high Bs(1.4T) and high thermal stability(>700°C). The magnetic properties are very sensitive to the volume fraction of HfC particles. After annealing at 700°C, Co-Hf-C films of the optimum composition consist of a mixture of fine fcc-Co and HfC grains. The grain size of fcc-Co is 10~15nm and that of HfC is 3~4 nm. The HfC particles are located at the grain boundaries of fcc-Co. HfC was found to crystallize before the fcc-Co, and the fine dispersion of these particles successfully inhibited the grain growth of the fcc-Co.**

I. INTRODUCTION

With the introduction of recording media of ever increasing coercivity (Hc), a corresponding enhancement in the gap field and sensitivity is necessary in order for the inductive head to operate at the higher frequency range. Thus, a high saturation flux density (Bs) and a high permeability (μ) are needed for head core materials. In fabricating Metal-In-Gap heads, thermal stability of the soft magnetic properties of the films is essential because they are subjected to high temperature (>500°C) during the glass bonding process.

Recently a number of high-Bs Fe-based or Co-based films with extremely fine grains have been investigated as suitable candidates as head materials [1]-[7]. These films showed good soft magnetic properties because the effective magnetic anisotropy is decreased by intergranular exchange interactions [8], [9]. Previously, we have found high μ, high Bs as well as good thermal stability in fine granular Co-based films (Co-M-C; M=Zr, Hf, Ta) [10]. It was thought that the M carbide may have prevented the Co grain growth. However, the grain size and distribution were not reported due to their fine nature. In this study, we investigate the microstructure of Co-Hf-C films which have shown better thermal stability than either Co-Zr-C or Co-Ta-C films, by transmission electron microscope (TEM), as well as by high resolution TEM (HRTEM).

II. EXPERIMENTAL PROCEDURE

Co-Hf-C films, 2μm thick, were deposited on partially crystallized glass substrates by reactive rf magnetron sputtering. The sputtering target was comprised of Hf chips placed on a pure Co target. CH_4 and Ar were introduced into the chamber for reactive sputtering. The total pressure was 1 Pa and the ratio of CH_4 /Ar was 1/5. The films were annealed in nitrogen gas for 30 minutes at 500 or 700°C. An in-plane rotating magnetic field (80kA/m, 40rpm) was applied during annealing. The composition of the film was measured by electron probe micro analyzer (EPMA). X-ray diffraction (Rigaku), TEM (Philips EM 420T) and HRTEM (JEOL JEM-4000EX) were used to study the structure of the films.

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III. RESULTS AND DISCUSSION

Table 1 shows the magnetic properties of the Co-Hf-C films after annealing at 700°C. We have studied two films with different compositions. Film A has the composition that shows the optimum soft magnetic properties and high thermal stability. On the other hand, film B has a higher Hf content and has lower μ and Bs than film A.

TABLE 1
Magnetic properties of Co-Hf-C films after annealing at 700°C for 30 minutes.

	Film A	Film B
Film Composition (at%)	$Co_{74}Hf_8C_{18}$	$Co_{66}Hf_{16}C_{18}$
Relative Permeability μ 5MHz	2000	600
Coercivity Hc (A/m)	40	120
Saturation Flux Density Bs (T)	1.4	1.0
Magnetostriction λ (x10^{-6})	-2	-3
Anisotropy Field (A/m)	400	200

Fig. 1 shows the x-ray diffraction spectra of film A and film B after annealing at 700°C. The peaks in these films correspond to those of bulk fcc-Co and HfC. The hcp-Co is not present in detectable amounts in these films. Film B shows larger HfC peaks because the Hf content of film B is greater than that of film A.

Fig. 2 (a) and (b) show the TEM bright field (BF) and dark field (DF) images for film A. The DF image was taken with the fcc-Co (111) and (200) rings. No DF images could be obtained using HfC rings because the volume fraction was too low. From the BF image, it can be seen that the grain size varies from 3 to 15 nm. However, the DF image reveals the grain size of fcc-Co to be around 10~15 nm. We can observe stacking faults and twin boundaries in fcc-Co, which could have formed during the annealing cycle during which the amorphous to crystalline transformation occurred.

Fig. 3 (a) shows a BF image of film B. Fig. 3 (b) and (c) show the DF images taken with the fcc-Co (111) and HfC (200) rings, respectively. For this film, the images from HfC

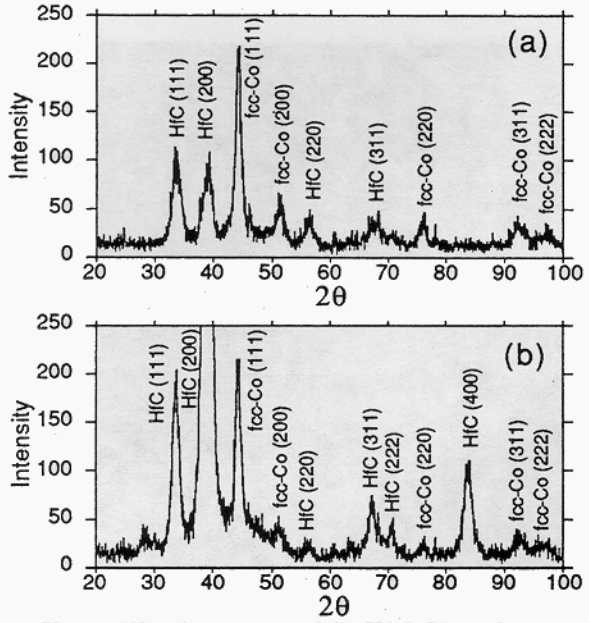

Fig. 1. X-ray diffraction spectra of Co-Hf-C films after annealing at 700°C for 30 minutes, : (a) film A and, (b) film B.

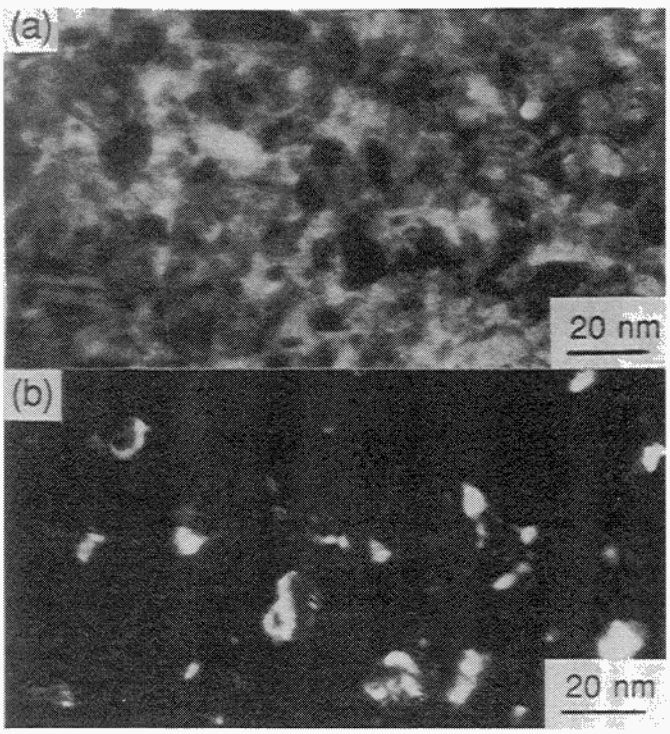

Fig. 2. TEM micrographs of Co-Hf-C film A after annealing at 700°C for 30 minutes, : (a) bright field image, (b) dark field image from fcc-Co (111) and (200).

rings could be obtained because of the higher volume fraction. From these images it can be seen that the grain size of fcc-Co is 10~15 nm and that of HfC is 3~10 nm. The shape of the HfC grains is equiaxed. The grain size of the fcc-Co in film B is similar to that of film A. Therefore, these results show that the additional HfC particles cause a reduction of the magnetic intergranular interaction between the fine fcc-Co grains, thereby lowering μ.

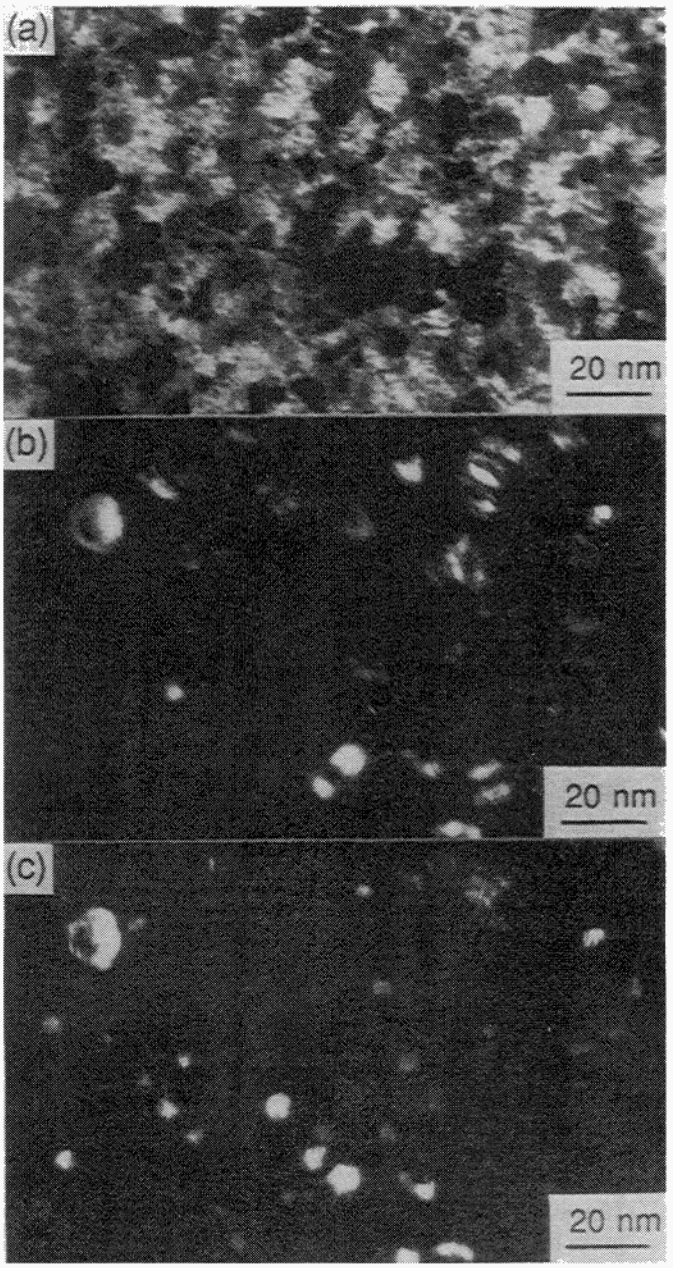

Fig. 3. TEM micrographs of Co-Hf-C film B after annealing at 700°C, : (a) BF image and, DF image, from (b) fcc-Co (111) and (c) HfC (200).

The structure of the as-deposited film and that annealed at 500°C were studied in order to understand how HfC particles prevent the grain growth of Co. Fig. 4 shows the x-ray diffraction spectra of the films with the same composition as film B in the as-deposited and annealed state. The as-deposited film has an amorphous structure. Only peaks of HfC were detected after annealing at 500°C. The low intensity and broadness of the Co peak show that Co is still amorphous. HfC particles were formed before Co because the free energy released during the formation of HfC is higher. The nucleation of HfC occurred uniformly in the films, resulting in a fine dispersions of nano size particles in the

film. This prevents the grain growth and coarsening of the subsequently crystallized fcc-Co.

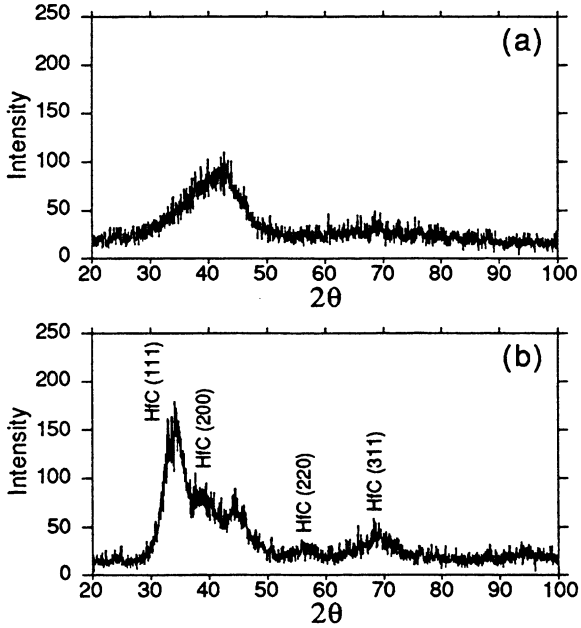

Fig. 4. X-ray diffraction spectra of Co-Hf-C films with the same composition as film B, : (a) as-deposited and, (b) after annealing at 500°C for 30 minutes.

We have also studied film A by HRTEM. Fig. 5 shows the atomic structure of a twined [110] fcc-Co grain. HfC particles can be found along the Co grain boundary. In this photograph only the {111} planes of the HfC particles are resolved. The (111) planes of one of the twin variants in the Co grain lines up with the (111) planes (white arrows) of a HfC grain, which indicates the heterogeneous nucleation and growth of Co on HfC. The subsequent grain growth occurred only in the upward direction because downward growth was arrested by the presence of HfC particles. As a result, the Co grain size is refined by this fine dispersion of HfC particles. However, an excessive amount of HfC particles along the Co grain boundary will reduce the intergranular exchange leading to a decrease in the permeability and an increase in the coercivity.

IV. CONCLUSION

We have carried out structural investigations on fine grain Co-Hf-C films. After annealing at 700°C, Co-Hf-C films of optimum composition consist of a mixture of fine fcc-Co and HfC grains. The grain size of fcc-Co is 10-15 nm and that of HfC is 3-4 nm. HfC particles were found to crystallize before fcc-Co. Therefore, this fine dispersion of HfC particles can prevent grain growth of the subsequently nucleated Co. In addition, the intergranular exchange coupling depends strongly on the volume fraction of HfC.

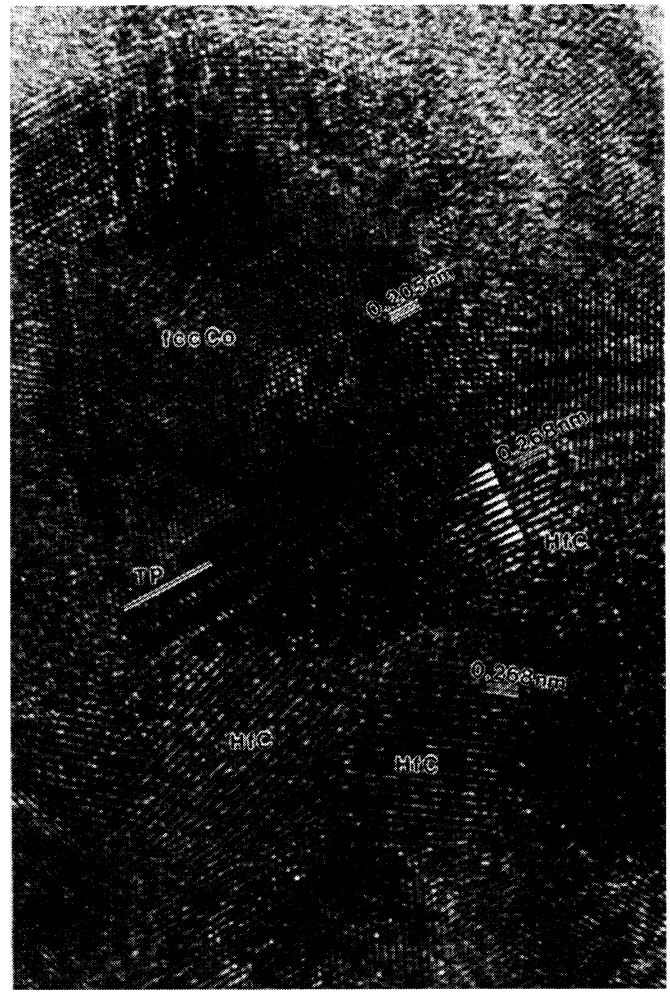

Fig. 5. HRTEM micrograph of Co-Hf-C film A after annealing at 700°C for 30 minutes, TP: Twin Plane.

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