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A Metallurgical Study on Sputtered thin Film Magnet of high $_iH_c$ Nd-(Fe, Co)-B alloy and Magnetic Properties

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Abstract Thin film magnet was fabricated by radio frequency magnetron sputtering using $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ alloy target and magnetic properties were investigated according to sputtering conditions from the metallurgical point of view. We could obtain the best preferred orientation of $Nd_2Fe_{14}B$ phase at substrate temperatures between 450℃ and 460℃ with the input power 150W, and thin films had the anisotropic magnetic properties. But, as the thickness of thin film increased, the c-axis orientation gradually tended to be disordered and magnetic properties also became isotropic. Just like Nd-Fe-B melt-spun ribbon, the microstructure of thin film magnet was consisted of very fine cell-shaped $Nd_2Fe_{14}B$ phase and the second phase along grain boundary. While, domain structure showed maze patterns whose magnetic easy axis was perpendicular to film plane of thin film.

It was concluded from these results that the perpendicular anisotropy in magnetization was attributed to the perpendicular alignment of very fine $Nd_2Fe_{14}B$ grains in thin film.

1. INTRODUTION

Recently, permanent magnetic materials that are composed of rare-earth metal and transition metal, such as $SmCo_5$, Sm_2Co_{17}, $Nd_2Fe_{14}B$, have been developed and attracting much attention[1~3]. Because of their high coercive force($_iH_c$) and energy product $((B \cdot H)_{max})$, they are used in the printer of computer, actuator of motive magnetic disk, special motor, various acoustic and communication device, and so on. However, they have always been using as bulk-shaped magnets.

Therefore, the development of thin film magnet which has high magnetic anisotropy without volume will be very attractive and important[4~6]. For example, firstly they can miniaturize various devices, secondly have superior magnetic properties different from bulk magnet, and thirdly they will be used as the new perpendicular magnetization recording media.

This study examined the magnetic properties and preferred orientation of Nd-(Fe·Co)-B thin film magnet according to various sputtering conditions from the standpoint of metallurgy.

2. EXPERIMENTAL PROCEDURE

This film magnet was fabricated by radio frequency (R.F) magnetron sputtering using $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ alloy target. The base pressure of sputtering was 4.1×10^{-6} Torr and the pressure during sputtering was held at 6.0×10^{-3} Torr in a high purity argon gas. The target was preliminary sputtered in order to eliminate oxide on target. After then we opened the shutter and fabricated thin film of 6mm in diameter. Thickness of thin film magnets was between 1 and 5㎛. The substrates used were silica glass and quartz, and the temperature of substrate ranged from room temperature to 500℃. Sputtering rate was controlled by target bias voltage and structure analysis was investigated by X-ray diffraction method(XRD). The magnetic properties of thin film were measured by vibrating sample magnetometer(VSM) whose maximum magnetic field was 20KOe. Magnetic domain pattern was observed by polarized optical microscope using SIGMAKER (Sigma Hi-Chemical Inc.) and by Kerr microscope at Yamasaki's lab. in Japan. Microstructure and composition analysis were studied by SEM, TEM, and electron probe micro-analysis(EPMA).

3. SESULTS AND DISCUSSION

3-1. Crystal Structure of Thin Film Magnet

Fig. 1 shows XRD patterns for thin film magnets sputtered at various substrate temperatures and annealed after sputtering. Fig. 1-(a) and (b) show XRD patterns of thin film which was sputtered at 420℃ for 10 and 30 minutes, respectively. They exhibit a hollow pattern of amorphous phase. The amorphous phase was also observed below the substrate temperatures of 420℃. However, when it was heated at 620℃ or over 620℃, they were crystallized clearly as shown in Fig. 1-(c). And, when it was raised the substrate temperature higher, for example above 450℃ and kept the sputtering time longer (for example from 30 minutes to 60 minutes at the same temperatures), thin films were not only crystallized directly without annealing, but also reflection peaks from many planes in addition to c-planes of thin film were observed. Fig. 1-(d), (e) show XRD patterns of thin film sputtered at 450℃ and 460℃ for 30 minutes, and Fig. 2 shows XRD patterns of thin film sputtered at 460℃ for 120 minutes. However, we couldn't observe so many c-plane peaks as the report of Yamasaki[7] and Yamashita[8]. We suppose that it was primarily due to the defect (for example, small cracks) of our target used for this study. These results show that the c-axis of thin film has oriented

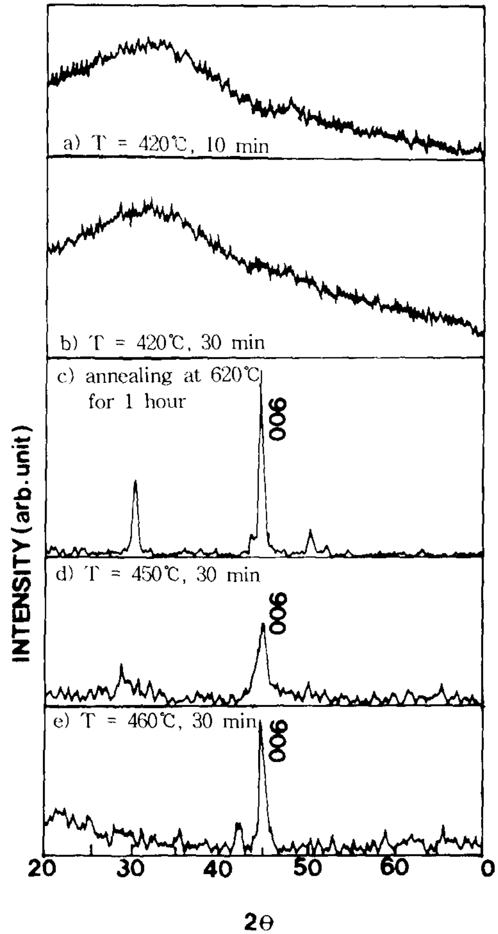


Fig. 1. XRD patterns of $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ sputtered thin film at various substrate temperatures.

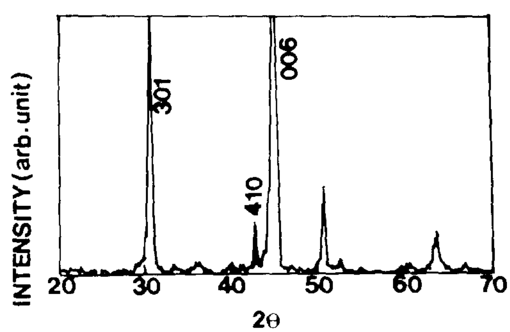


Fig. 2. XRD pattern of $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ thin film sputtered at 460℃ for 120min.

perpendicular to the film plane, but as sputtering time increases, that is, as the thickness of thin film magnet increases, c-axis orientation tended to be disordered gradually as Yamasaki has reported[7]. This means that the alignment of c-axis magnetization perpendicular to the film plane in Nd-(Fe·Co)-B thin film magnet could be possible when we have the optimum substrate temperature and sputtering time, as discussed in the latter magnetic properties.

3-2. Magnetic Properties of $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ Thin Film

The hysteresis loops of thin film measured perpendicular and parallel to the film plane were shown in Fig. 3 and 4. Fig. 3 shows hysteresis loops of thin film sputtered at different substrate temperatures, while Fig. 4 shows hysteresis loops for different sputtering time, that is, the effect of film thickness on the hysteresis loops of $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ thin film magnet.

As the former(Fig. 3) shows, although their intrinsic coercive force are not so large as expected, the magnetization measured perpendicular to film plane was very large compared to the parallel one. In other words, the thin films sputtered near 450℃ had the anisotropic

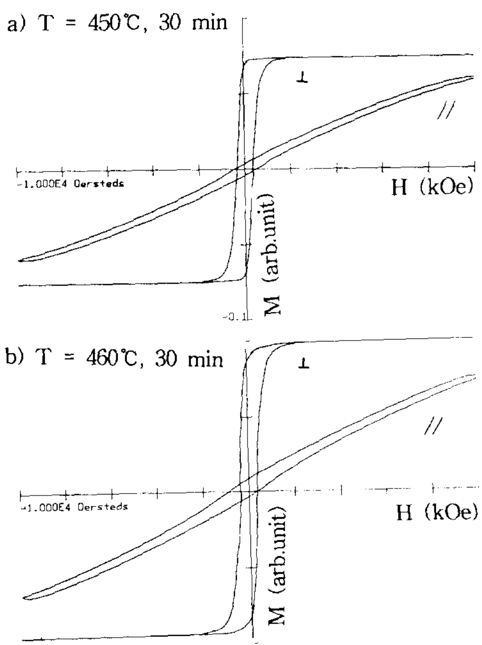


Fig. 3. Hysteresis loops of thin film sputtered at different substrate temperatures.

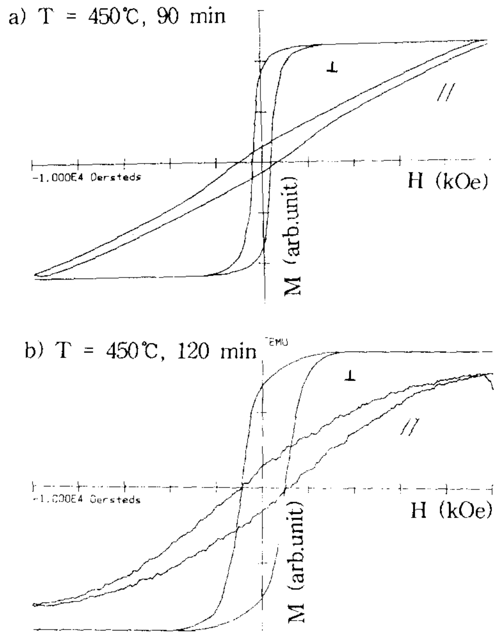


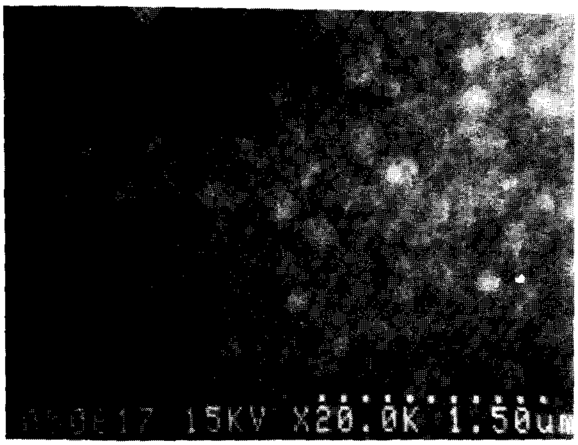
Fig. 4. Hysteresis loops of thin film sputtered for various sputtering time at 450℃.

magnetic properties, but it was noted in the latter that the anisotropic magnetic properties gradually changed into isotropic with the increase of film thickness. Relating these results to the previous c-axis orientation, it was thought that the anisotropic magnetic properties above are associated with the texture of thin film structure.

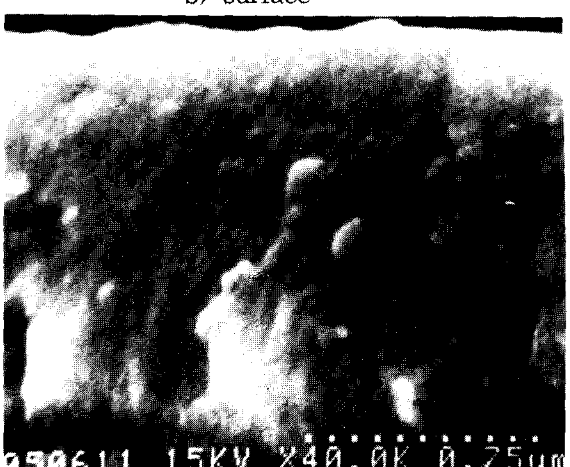
3-3. Microstructure and Domain Structure of $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ Thin Film Magnet

Fig. 5-(b), Fig. 5-(a) and 5-(c) are SEM micrographs for the surface and the perpendicular cross section of thin films which were fabricated by sputtering $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ magnet alloy at indicated conditions. It was noted from these SEM micrographs that microstructure of thin film magnet was consisted of very fine $Nd_2Fe_{14}B$ phase whose grain size was 20～50nm and a certain of the second phase along grain boundary. That was just like the microstructure of Nd-Fe-B melt-spun ribbon magnets. While, the grain structure in the cross section of thin films was like Fig. 5-(b). They had the very interesting

a) cross-section
T = 450℃, 90 min



b) surface



c) cross-section
T = 460℃, 60 min

Fig. 5. SEM micrographs of surface and perpendicular cross-section of $Nd_{13}(Fe\cdot Co)_{70}B_{17}$ sputtered thin film.

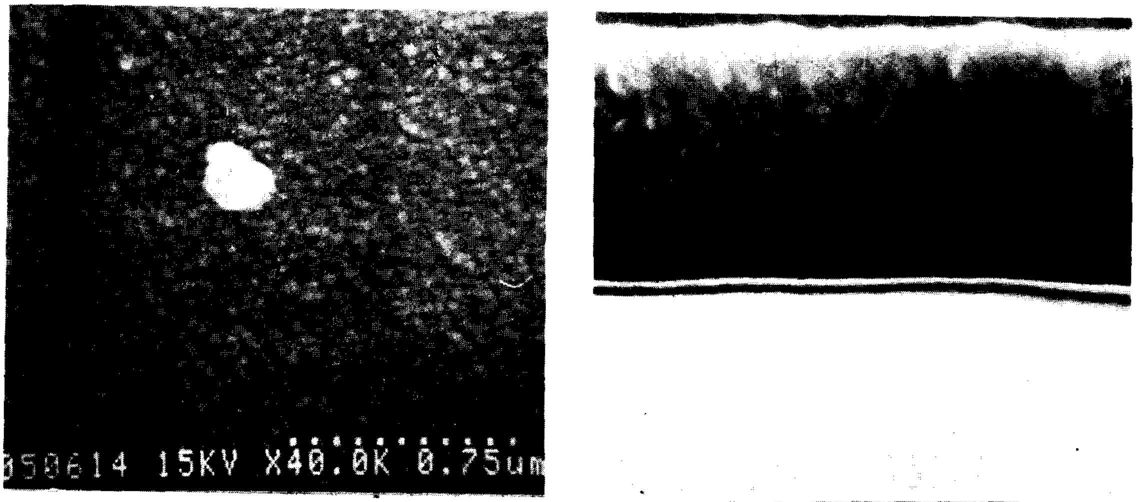
columnar structure running from surface to the interface between thin film and substrate. Their appearances are clearly related to the domain structures of thin film magnet which were shown in the latter. Fig. 6-(a) and (b) shows the same as microstructural appearences observed in the thin films which were sputtered at 450℃ for 90 minutes and at 460℃ for 60 minutes. That is, thin films which had the most superior anisotropic properties showed the clearer columnar structure.

We also tried to observe the microstructure of thin film magnet using transmission electron microscope (TEM), but TEM studies of the thin films sputtered were not fully successful because it was difficult to remove the films from substrate.

Fig. 7 shows a part of TEM micrographs of thin film sputtered $Nd_{13}(Fe\cdot Co)_{70}B_{17}$ magnet alloy at 460℃ for 60min. They also showed that the microstructure of $Nd_{13}(Fe\cdot Co)_{70}B_{17}$ thin film were just the same as that of the Nd-Fe-B melt-spun ribbon, but grain boundary phase was too complex to reveal. This observation is very important because we may presume the coercive mechanism of Nd-Fe-B thin film magnet, but further studies are to be required to elucidate this problem.

Fig. 8 are micrographs showing the magnetic domains of $Nd_{13}(Fe\cdot Co)_{70}B_{17}$ thin film whose c-axis orientation might be excellent.

Fig. 8-(a) was observed by polarized optical microscope using SIGMAKER and Fig. 8-(b) by a Kerr effect microscope. They showed the maze patterns. It is generally known that when the magnetic easy axis oriented perpendicular to the crystal surface, maze pattern could be observed. Now, relating the previous c-axis orientation of very fine $Nd_2Fe_{14}B$ grains and microstructural appearances to the domain patterns, it may be concluded that the anisotropic magnetic properties of thin films are clearly attributed to the perpendicular magnetization of c-axis in this films and the isotropical magnetic properties observed with the increase of thin films



surface

cross-section

Fig. 6. SEM micrographs of surface and perpendicular cross-section of the thin film

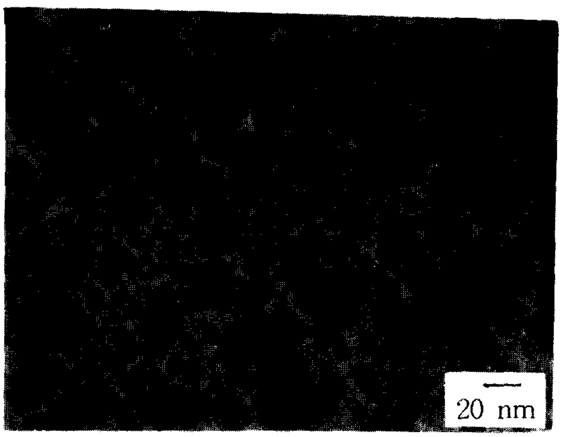
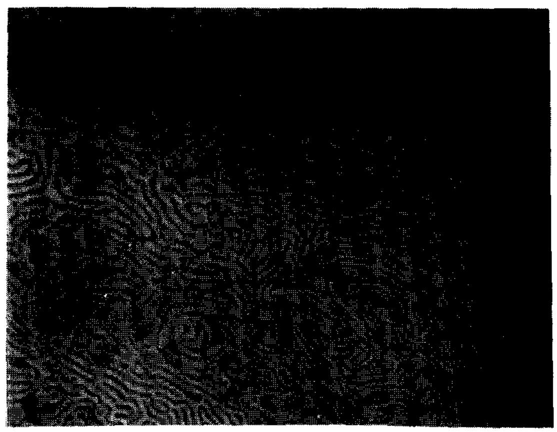
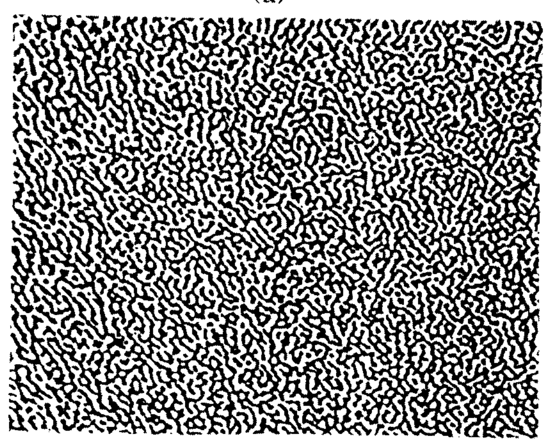


Fig. 7. TEM micrographs of thin film surface sputtered at 460℃ for 60 min.



(a)



(b)

Fig. 8. Magnetic domain patterns of film magnet observed by a) Bitter method b) Kerr microscope (Photographed at Prof. Yamasaki Lab.)

thickness are attributed to the gradual disorder from perpendicular orientation to plane orientation of c-axis in thin film magnet.

4. CONCLUSION

1) In case of $Nd_{13}(Fe\cdot Co)_{70}B_{17}$ magnetic alloy, we could obtain thin film magnet having the best c-axis orientation at input power 150W and substrate temperature between 450℃ and 460℃.

2) The c-axis orientation as well as magnetic properties of Nd-Fe-B based thin film manget changed as the thickness of thin

film magnet increased. (for instance, from perpendicular anisotropy to plane anisotropy)

3) The microstructure of thin film magnet composed the same fine cell structure as melt-spun Nd-Fe-B ribbons made up by rapidly quenching, but the secondary phase along grain boundary was too complicated to reveal. Their grain size was 20~50 nm.

4) The domain structure of $Nd_{13}(Fe \cdot Co)_{70}B_{17}$ thin film magnet showed maze patterns whose magnetic easy-axis was perpendicular to film plane and the change of domain structure according to the thickness of film and substrate temperature was consistant with the change of c-axis orientation.

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