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Sm-Fe-N Thick Film Magnets Fabricated by Aerosol Deposition Method

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This paper describes the possibility of the aerosol deposition (AD) method for the fabrication of Sm-Fe-N thick films. The thickness (*d*) of AD films reached to 190 μm after the deposition with gas flow rate (*gfr*) = 6 l/min and deposition time (*t*) = 12 min. The deposition rate was estimated as 15-19 μm/min. The Sm-Fe-N AD films showed the remanence (B_r) of 0.36-0.42 T, which were 54-63 % of that of host powder (0.67 T). The coercivities ($\mu_0 H_{cJ}$) increased from 1.16 T to 1.69-1.79 T because of grain refinement during the deposition. The effect of applied field during AD method was also investigated. Applying magnetic fields of 0-0.19 T in plane, the remanence measured along the applied field increased with increasing the magnetic field. The maximum value of remanence was 0.54 T, which was 29% higher than that without the applied field (0.42 T). It is concluded that the c-axis has a tendency to align along the magnetic field and the AD method is effective for the fabrication of thick film magnets.

Key words: aerosol deposition, anisotropy, magnetic field, samarium iron nitrogeon, thick film magnet

1. INTRODUCTION

Rare earth magnets with thickness less than 300 μm are strongly demanded in the trend of downsizing motors and electromagnetic devices. A mechanical process is one of the simple methods to reduce the thickness of bulk rare earth magnets. However, the influence of defects introduced at the surface leads to decrease magnetic properties. On the other hand, sputtering and pulsed laser deposition (PLD) methods are convenient to obtain film magnets [1-4]. However, their deposition rate is not so sufficient for manufacturing film magnets.

The aerosol deposition (AD) is one of the attractive methods as a film fabrication process [5]. In our previous papers [6-8], we applied this method for the fabrication of Sm-Fe-N thick film magnets and high coercivity was reported. In this paper, our recent investigations about Sm-Fe-N AD films including the relationships among AD conditions, magnetic properties and microstructure were described. In addition, the effect of applied field during AD and the possibility of anisotropic Sm-Fe-N film magnets were investigated.

2. EXPERIMENTAL PROCEDURE

Figure 1 shows the schematic illustration and conditions of AD method. The Sm-Fe-N powder was provided from the Sumitomo Metal Mining Co. Ltd.,. The principle and details of the AD method were described in our previous paper [7]. A helium gas was used as a carrier gas and the gas consumption (gas flow rate: *gfr*) was varied from 2 to 6 l/min. The deposition time (*t*) was also changed from 2 to 12 min. The substrates used in this investigation were SiO_2 and brass with the size of 10 mm x 10 mm x 1 mm. A metal mask made of stainless steel or cellophane with the opening size of 8 mm x 8 mm, was also used. They were placed at a distance of 10 mm from the nozzle, and were maintained at room temperature.

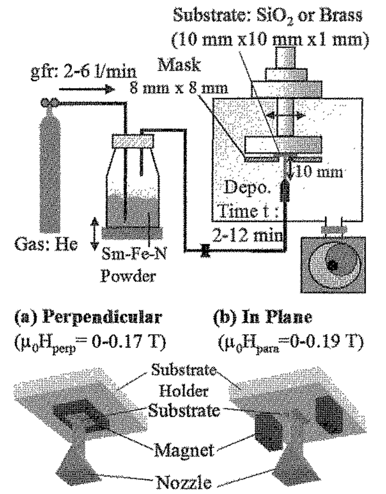

Fig.1 Schematic illustration and conditions of AD method in this study.

In order to apply a magnetic field during the deposition, one or two Nd-Fe-B permanent magnets were placed near the substrate. In the case of applying a magnetic field perpendicular to the film plane ($\mu_0 H_{perp}$), one Nd-Fe-B magnet was placed behind the substrate. The $\mu_0 H_{perp}$ was changed from zero to 0.17 T by changing the distance between the magnet and the substrate. Similarly, in the case of applying a field in plane ($\mu_0 H_{para}$), two Nd-Fe-B magnets were placed facing each other across the substrate. The $\mu_0 H_{para}$ was controlled in the range of 0-0.19 T by changing the distance between the two magnets.

The area and thickness of AD films were measured by a surface profilometer, which were used for the

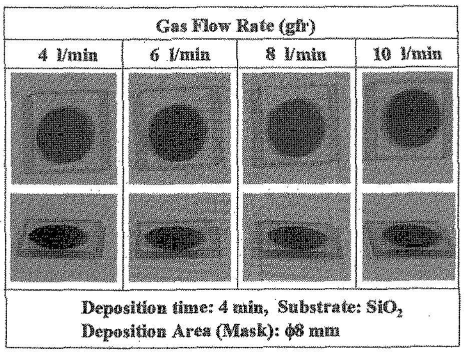

Fig.2 Photographs of the Sm-Fe-N AD films.

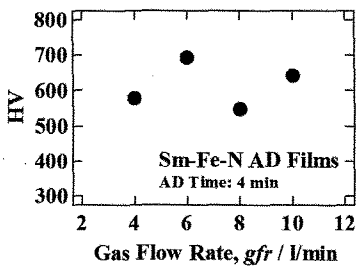

Fig.3 Vickers hardness of the SmFe-N AD film deposited for 4 min.

calculation of film volume and film density. The magnetic properties were measured by a vibrating sample magnetometer (VSM) after applying a pulsed field around 4 T. The microstructures were observed using X-ray diffraction (XRD) and scanning electron microscopy (SEM) techniques.

3. RESULTS AND DISCUSSION

3.1 Deposition rate

Figure 2 shows the photographs of Sm-Fe-N AD films in as-deposition state. From these photographs, there is no evidence of peeling off and thick films were obtained. Figure 3 shows the micro-vickers hardness of the AD films deposited under the condition of *gfr* = 4-10 l/min for 4min. The *HV* values were around 500 to 700 and these films showed relatively high hardness.

Figure 4 shows variation in thickness (*d*), density (ρ) and relative density (ρ_r) of the Sm-Fe-N AD films deposited for 4 min as a function of the gas flow rate (*gfr*). Under the conditions of *gfr* = 2 and 4 l/min, the *d* was not large. However, the *d* increased with increasing *gfr*, and the maximum thickness (77 μm) was obtained at *gfr* = 6 l/min. From this result, the deposition rate was calculated as 19 μm/min, which was one order higher than that of conventional PLD method. The ρ and ρ_r showed almost the same tendency as *d*, and the values were 4.9-5.9 g/cm^3 and 64-77 %, respectively.

Figure 5 shows the variations in *d*, ρ and ρ_r of the Sm-Fe-N AD films fabricated with *gfr* = 6 l/min as a function of the deposition time (*t*). The *d* increased with increasing *t* and the maximum thickness of 190 μm was obtained with the sample deposited for 12 min. The deposition rate was calculated as around 15 μm/min. The ρ was 5.4-6.2 g/cm^3, which corresponded to the ρ_r of 71-81 %.

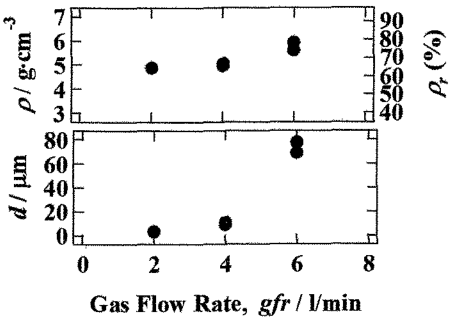

Fig. 4 Variation of thickness (d), density (ρ) and relative density (ρ_r) of Sm-Fe-N AD films as a function of gas flow rate (gfr)

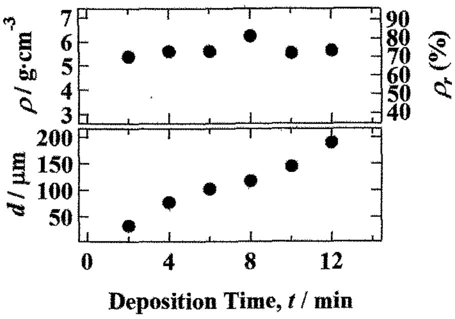

Fig. 5 Variation of thickness (d), density (ρ) and relative density (ρ_r) of Sm-Fe-N AD films as a function of deposition time (t).

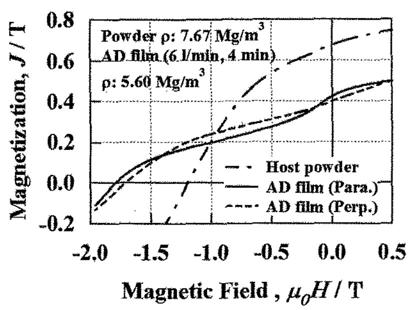

Fig. 6 The demagnetization curve of the Sm-Fe-N AD film deposited with gfr = 6 l/min for 4min in comparison with that of the host powder.

3.2 Magnetic properties

Figure 6 shows the demagnetization curves of the Sm-Fe-N host powder and the AD film deposited with *gfr* = 6 l/min for 4 min. The demagnetization curve of the AD film was measured along the direction parallel (para.) or perpendicular (perp.) to the film plane. The Sm-Fe-N AD film exhibited lower remanence (B_r) and higher coercivity (μ_0H_{cJ}) than the host powder. In addition, one inflection was observed in the demagnetization curves, which suggested the existence of a soft magnetic phase. Figure 7 summarizes magnetic properties of the AD films deposited with *gfr* = 2-6 l/min and *t* = 4 min. The B_r values were in the range of 0.36-0.42 T, which were 54-63 % of that of the Sm-Fe-N host powder (0.67 T). The μ_0H_{cJ} increased from 1.16 T to 1.69-1.91 T by the deposition.

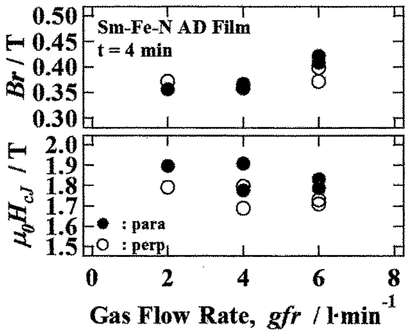

Fig.7 Magnetic properties of Sm-Fe-N AD films deposited with gfr = 2-6 l/min for 4min.

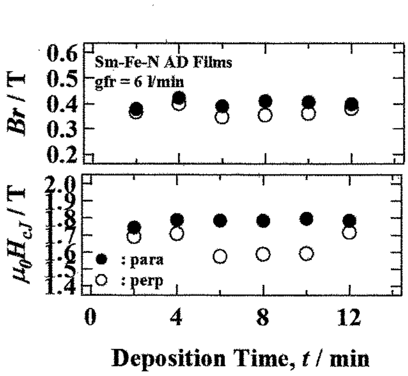

Fig. 8 Magnetic properties of the Sm-Fe-N AD films prepared under the conditions of gfr=6 l/min and t= 2-12 min.

Figure 8 shows the magnetic properties of the Sm-Fe-N AD films deposited under the conditions of gfr = 6 l/min and t = 2-12 min. The B_r and μ_0H_{cJ} were not so changed by the deposition time. The B_r was in the range of 0.38-0.42 T, which was 56-63 % of that calculated in the host powder. However, the AD films measured along the direction parallel to the film plane also exhibited high coercivity of 1.74-1.79 T.

3.3 Microstrucutre

In order to investigate the microstructure of Sm-Fe-N AD films, SEM observations were carried out. Figure 9 shows SEM photographs taken from the Sm-Fe-N host powder (a), surface (b) and cross section (c) of the AD film deposited by gfr = 6 l/min and t= 4 min. The host powder exhibited a flake like shape with an average size of 1.94 μm. From Fig. 9 (b), the surface of the AD film was not so flat, however, small particles of 0.2-0.4 μm, which were smaller than the average powder size of the host powder, were observed. Figure 9 (c) revealed that the shape of particles seemed to be ellipse and that the densification proceeded. The average grain size derived from this photograph was about 0.32 μm. It can be concluded that the grain refinement occurred during the AD, which resulted in a high coercivity.

Figure 10 shows XRD patterns of the Sm-Fe-N host powder and the AD film deposited with gfr = 6 l/min and t = 4 min. Both samples were mainly composed of the $Sm_2Fe_{17}N_x$ compound, however, small amount of α-Fe phase was observed in the AD film. This α-Fe phase influenced the inflection that observed in Fig. 6. From the XRD patterns, the ratio of X-ray peak intensity between (006) and (033) of the $Sm_2Fe_{17}N_x$ compound ($I_{(006)}/I_{(033)}$) was calculated. The AD film showed the ratio of 0.80, which was higher than that in the host powder (0.33). It can be said that the c-axis has a tendency to align along the direction perpendicular to the film plane.

3.4 Anistropic Films

In order to fabricate anisotropic films, the effect of applied field were investigated. In the case of applying magnetic field along the direction perpendicular to the film plane (μ_0H_{perp}) during the AD method, the density of the AD film decreased with increasing μ_0H_{perp} because of the magnetic cohesion. It resulted in the decrease in remanence and magnetic properties. In next, magnetic fields were applied along the direction parallel to the film plane (μ_0H_{para}) during the AD method. Figure 11 shows the variation of d, ρ and ρ_r of the Sm-Fe-N AD films with respect to the μ_0H_{para}. The d decreased with increasing μ_0H_{para}. However, the density was not so changed and was around 5.6 g/cm^3.

Figure 12 shows the demagnetization curves of the Sm-Fe-N AD film deposited without or with μ_0H_{para} of 0.19 T. Both of them were measured along μ_0H_{para}. The film deposited with μ_0H_{para} exhibited higher B_r.

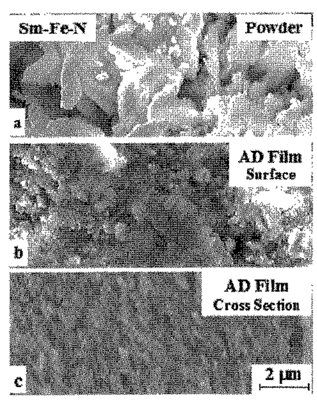

Fig.9 SEM photographs taken from the surface (b) and cross section (c) of the AD film prepared with gfr = 6 l/min and t= 4 min, in comparison with that of host powder (a).

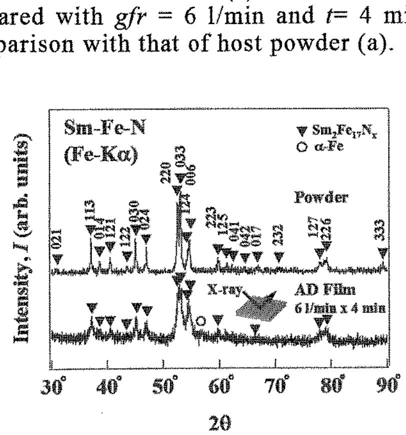

Fig.10 The XRD pattern of the Sm-Fe-N AD film prepared with gfr = 6 l/min and t = 4 min in comparison with that of host powder.

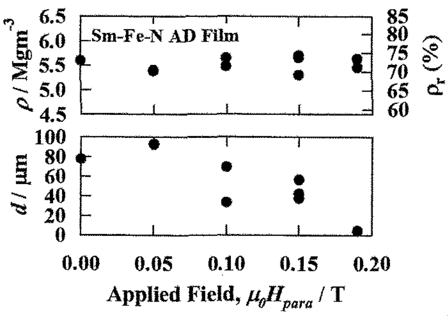

Fig. 11 Variation of thickness (d), density (ρ) and relative density (ρ_r) of the Sm-Fe-N AD films deposited with μ_0H_{para}.

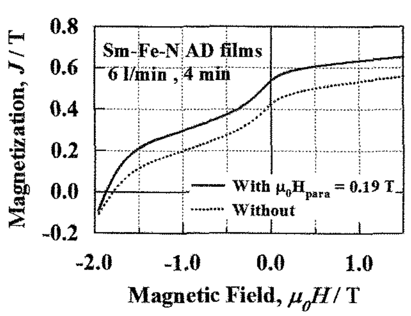

Fig. 12 The demagnetization curves of Sm-Fe-N AD film deposited with or without μ_0H_{para}.

Figure 13 summarizes B_r of the Sm-Fe-N AD films deposited with μ_0H_{para} of 0-0.19 T. The coercivities (μ_0H_{cJ}) measured along the direction parallel to the μ_0H_{para} (para. (in plane)) were 1.79-1.87 T. They were higher than those measured along the direction perpendicular to the μ_0H_{para} (perp. (in plane) and perp.). The B_r measured along the μ_0H_{para} direction (para. (in plane)) increased with increasing the μ_0H_{para}. The maximum value of B_r was 0.54 T, which was 29% larger than that of without μ_0H_{para} (0.42 T). On the other hand, the B_r measured along the direction perpendicular to the μ_0H_{para} (perp. (in plane) and perp.) seemed to relatively decrease with increasing the μ_0H_{para}. Figure 14 showed the variation of $I_{(006)}/I_{(033)}$ as a function of μ_0H_{para}. The value decreased with increasing μ_0H_{para}, which suggested the c-axis of the $Sm_2Fe_{17}N_x$ compound aligned in plane. Namely, the c-axis has a tendency to alogn along the magnetic field direction and the anisotropy increased with increasing the field.

Finally, it is concluded the AD method is very effective method for the fabrication of thick film magnets with high deposition rate. In addition, anisotropic film magnets are considered to be prepared by applying magnetic field during the AD method.

4. ACKNOWLEDGEMENT

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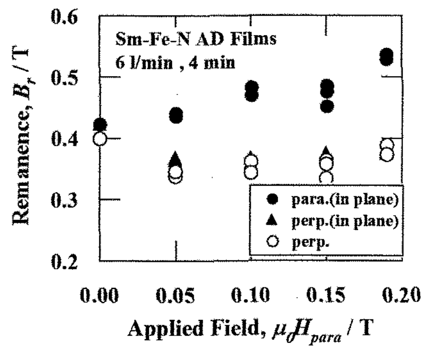

Fig. 13 The B_r of the Sm-Fe-N AD films deposited with μ_0H_{para} of 0-0.19 T. The films were measured in parallel (para. (in plane)) or perpendicular (perp. (in plane) and perp.) to the direction of μ_0H_{para}.

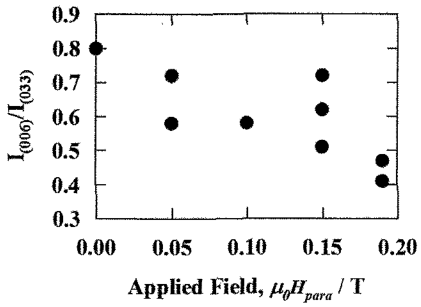

Fig. 14 The variation of $I_{(006)}/I_{(033)}$ taken from the Sm-Fe-N AD films deposited with μ_0H_{para} of 0-0.19 T.

5. REFERENCES

[1] F. J. Cadieu, T. D. Cheng, and L. Wickramasekara, J. Magn. Magn. Mater., 54-57, 535-536, (1986).
[2] S. Yamashita, J. Yamasaki, M. Ikeda, and N. Iwabuchi, J. Appl. Phys., 70, 6627-6629, (1991).
[3] M. Nakano, S. Tsutsumi and H. Fukunaga, IEEE Trans. Magn., 39, 2863-2865, (2003).
[4] D. J. Sellmyer, J. Zhou, Y. Liu, R. Skomski, Proc. 17th International Workshop on Rare Earth Magnets and Their applications, ed. by G. C. Hadjipanayis and M. J. Bonder, (Rinton Press, Princeton, New Jersey, 2002) pp. 428-437.
[5] J. Akedo and M. Lebedev, Jpn. J. Appl. Phys., 40, 5528-5532, (2001).
[6] S. Sugimoto, T. Maeda, R. Kobayashi, J. Akedo, M. Lebedev and K. Inomata, IEEE Trans. Magn., 39, 2986-2988, (2003).
[7] S. Sugimoto, J. Akedo, M. Lebedev, and K. Inomata, J. Magn. Magn. Mater., 272-276, E1881-E1882, (2004).
[8] S. Sugimoto, T. Maki, T. Kagotani, J. Akedo and K. Inomata, J. Magn. Magn. Mater., (accepted).

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