Nd-Fe-B thick-film magnets prepared by high laser energy density

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An observation of microstructure revealed that a laser-irradiated Nd-Fe-B target under the laser energy density (LED) above 10 J/cm^2 enabled us to prepare isotropic nano-composite thick-film magnets with good magnetic properties due to a dispersed α-Fe + Nd-Fe-B structure. The formation of the structure is attributed to the etching process of a target during a deposition. Moreover, the control of composition in the nano-composite film enhanced *(BH)*$_{max}$ value up to approximately 130 kJ/m^3.

***Index Terms*—Nd-Fe-B, PLD (Pulsed Laser Deposition), laser energy density, droplet**

I. INTRODUCTION

Many researchers have demonstrated miniaturized devices comprising anisotropic Nd-Fe-B film magnets.[1]-[3] Recently, our group reported isotropic Nd-Fe-B+α-Fe nano-composite film magnets prepared by a PLD (Pulsed Laser Deposition) method using the small spot size of laser beam less than 1 mm together with LED (Laser Energy Density) ≧ 10 J/cm^2 in order to apply the films to a multi-polarly magnetized rotor for a miniaturized cylindrical motor.[4,5] The deposition rate of a previously-reported process using the large spot size of several mm by taking account of a defocused laser beam on the surface of a target (LED ≦ 1 J/cm^2) reached 90 μm/h [6] and the film thickness increased up to 1300 μm.[7] On the other hand, the above-mentioned process using LED ≧ 10 J/cm^2 had a relatively low deposition rate of 20- 40 μm/h and the good magnetic properties of Br ≧ 0.9 T, Hc ≧ 300 kA/m, and *(BH)*$_{max}$ ≧ 80 kJ/m^3, respectively, could be confirmed only within the thickness range from 10 to 20 μm. [5]

In this study, an observation on the microstructure of a Nd-Fe-B nano-composite thick-film prepared by LED above 10 J/cm^2 was carried out to examine the origin for obtaining good magnetic properties. In order to investigate the process to form the above-mentioned microstructure, the relationship between magnetic properties and deposition time was also studied. Furthermore, we succeeded in an enhancement in *(BH)*$_{max}$ by taking account of the composition for the nano-composite films.

II. EXPERIMENTAL PROCEDURE

A rotated target with the composition of $Nd_{2.4}Fe_{14}B$ was ablated by an Nd-YAG pulse laser (wave length = 355 nm). The films deposited on each Ta substrate were fabricated in the vacuum atmosphere of 10^{-5} Pa [8]. A laser energy density (LED) varied by a spot size of laser beam which could be controlled by moving the distance between a focal lens and a target intentionally. The range of LED above 10 J/cm^2 was used under fixing the laser power of 4 W which was measured with a power meter in front of the entrance lens of a chamber. The deposition rate became from 20 to 40 μm/h by using the distance of 10 mm between a target and a substrate. All the as-deposited films thicker than 10 μm were crystallized by a pulse annealing (PA) method in the vacuum atmosphere of 2-5×10^{-5} Pa. [9]

The magnetic properties of the annealed samples were measured with a vibrating sample magnetometer (VSM) under applied maximum magnetic field of 2.5 T after magnetization under a pulsed magnetic field of 7 T. In the results, in-plane magnetic properties were only shown because all the films had isotropic magnetic properties. The film thickness was measured with a micrometer and the composition of each film was analyzed with an energy dispersive X-ray spectrometry (EDX). The microstructure was observed with a Transmission Electron Microscope (TEM).

III. RESULTS AND DISCUSSION

A. Magnetic properties and Microstructure of Nd-Fe-B thick-film magnets prepare by LED ≧ 10 J/cm^2

Figure 1 shows *(BH)*$_{max}$ values of Nd-Fe-B thick-film magnets prepared by using LED ≦ 1 J/cm^2 and LED ≧ 10 J/cm^2, respectively. Although the Nd contents of each film were almost the same range of between 8 and 10 at %., the average *(BH)*$_{max}$ value of the samples prepared by high LED was higher than that of low LED-made ones by approximately 20 kJ/m^3. In order to investigate the mechanism for obtaining superior *(BH)*$_{max}$ due to the increase in LED as shown in Fig.1, the microstructure of a sample prepared by LED ≧ 10 J/cm^2 was observed. The sample was prepared by using the deposition time of 30 min. As displayed in Fig. 2, a dispersed α-Fe + Nd-Fe-B structure could be obtained and it was confirmed that the average diameter of α-Fe grains was approximately 20 nm,

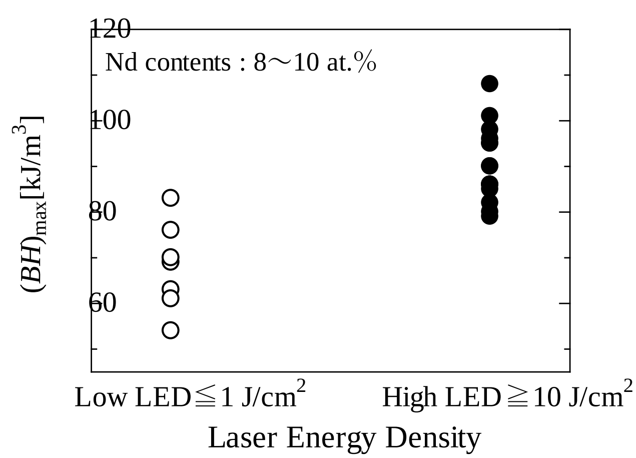

Fig. 1 *(BH)*$_{max}$ of Nd-Fe-B thick-film magnets prepared by different values of LED. The Nd content range of each film prepared by LED less than 1 J/cm^2 and higher than 10 J/cm^2, respectively, was almost the same as from 8 to 10 at %., however the value of *(BH)*$_{max}$ for the samples prepared by high LED was superior.

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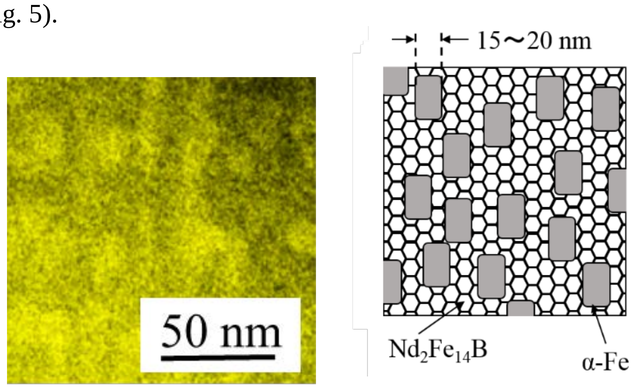
which is one of optimum conditions for a nano-composite magnet considering an exchange interaction length. Namely, usage of LED above 10 J/cm^2 enabled us to fabricate an isotropic nano-composite thick-film magnet with good magnetic properties due to the ideal microstructure.

In order to investigate the mechanism to form the microstructure, the relationship between various properties and deposition time was examined. The extension of continuous deposition time up to 180 min. was carried out under LED ≧ 10 J/cm^2 as seen in Fig. 3. The thickness increased linearly up to 120 min. as the deposition time prolonged and the drastic enlargement in the thickness at the deposition time of 150 min. could be observed. It was found that the phenomenon was attributed to the large amount of emitted droplets from a target. (see Fig. 4) In the case of LED ≦ 1 J/cm^2, the use of droplets was a key technology to obtain deposition time became over 10 h (Total thickness : 1300 μm).[7] On the other hand, as a sample was fabricated by the maximum deposition rate of approximately 90 μm/h[6] and the deposition rate was almost the same even if the using LED ≧ 10 J/cm^2, the droplets were explosively emitted due to the progress of erosion a Nd-Fe-B target after a continuously long deposition up to 180 min(see Fig. 5).



(a) TEM observation (b) Schematic diagram of Fig. 2 (a)

Fig. 2 Photograph and schematic diagram of microstructure for an isotropic nano-composite thick-film magnet with a dispersed α-Fe + Nd-Fe-B structure. The sample was prepared by using LED ≧ 10 J/cm^2 under the deposition time of 30 min.

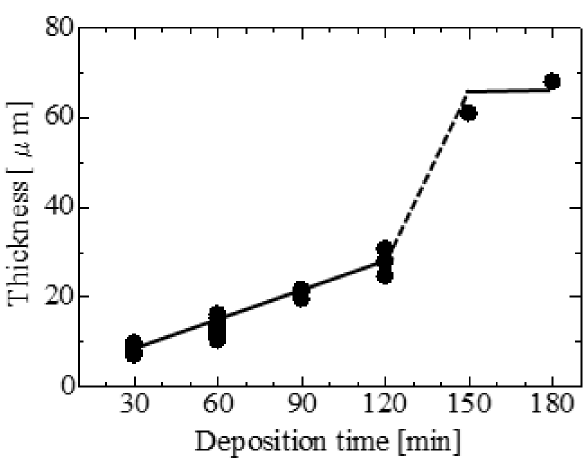
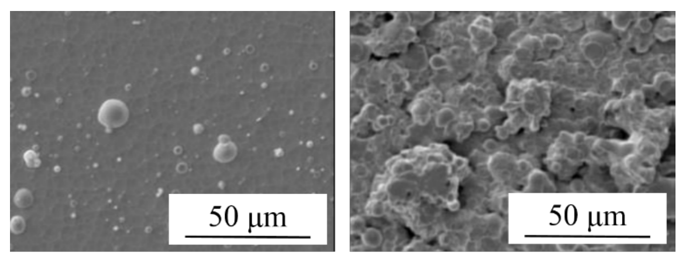


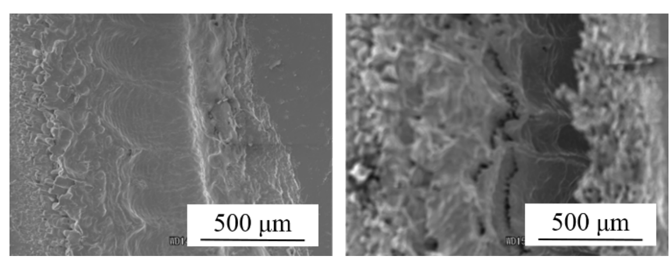
Fig. 3 Thickness of film magnets prepared by using LED ≧ 10 J/cm^2 as a function of deposition time. The drastic increase in the thickness occurred at the deposition time of 150 min.

In the case of deposition time of 180 min., the target was roughly etched and resultantly large amounts of droplets were emitted. Figure 6 shows the magnetic properties of the films plotted in Fig. 3. The average values of remanence and coercivity showed approximately 1.0 T and 350 kA/m, respectively, up to 120 min., and the remanence values extremely dropped under the prolongation of deposition time. The relative large variation of the remanence of the film deposited under 30min together with the coercivity of the film deposited under 60 min. is considered to be attributed to the fluctuation of Nd contents in each film as shown in Fig. 7. In addition, the measurement in the composition of surface area suggests that almost droplets had Nd-rich composition compared with stoichiometric one. We also considered that the erosion of a target due to the prorogation in the deposition time leaded to increase the size and/or numbers of droplets. Since the Nd content of each droplet showed almost the same value compared to that of a $Nd_{2.6}Fe_{14}B$ target, it was, therefore, considered that the emitting mechanism of the deposition time above 150 min. became similar to that of LED ≦ 1 J/cm^2. These results suggest that usage of a small spot size of a laser beam (LED ≧10 J/cm^2) within an optimum deposition time enabled us to obtain isotropic nano-composite thick-film magnets with good magnetic properties due to the-above mentioned microstructure.



(a) 60 min. (b)180 min.

Fig. 4 Surface morphology of film magnets prepared by using LED ≧ 10 J/cm^2 at the deposition times of 60 and 180 min. respectively. A large amounts of emitted droplets from a target could be observed in the sample deposited for 180 min.



(a) 60 min. (b) 180 min.

Fig. 5 Surface observation of each target ablated by LED ≦ 10 J/cm^2 for 60 and 180 min., respectively. The numbers and/or size of emitted droplets is relatively small because the target was delicately etched.

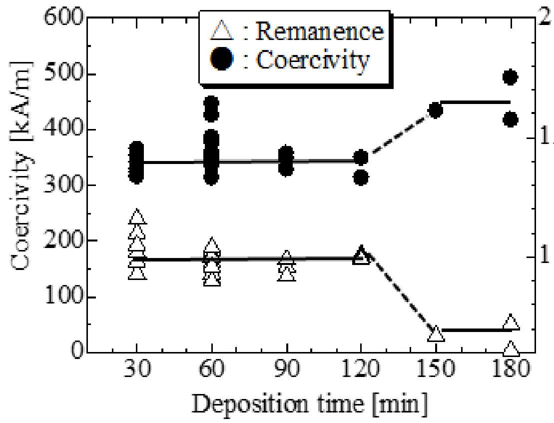


Fig. 6 Magnetic properties for Nd-Fe-B thick film magnets prepared by using LED ≧ 10 J/cm^2 as a function of deposition time. Remanence and coercivity showed constant values in the deposition time range from 30 to 120 min.

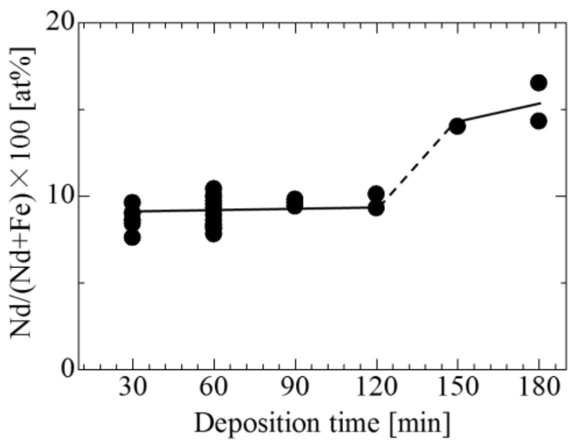


Fig.7 Nd contents of film magnets prepared by using LED ≧ 10 J/cm^2 as a function of deposition time. The amount of Nd extremely increased in the samples deposited for 150 and 180 min. due to the increase in the numbers and/or size of droplets.

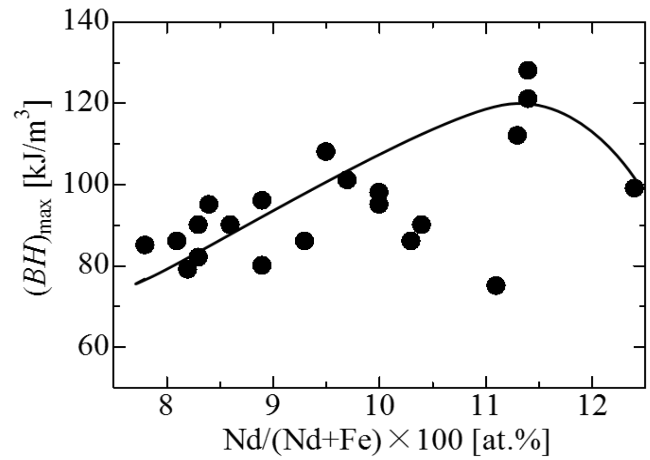


Fig. 8 $(BH)_{max}$ of Nd-Fe-B thick-film magnets prepared by LED≧10 J/cm^2 as function of Nd contents. The value of $(BH)_{max}$ increased with increasing Nd contents up to approximately 11.5 at. %.

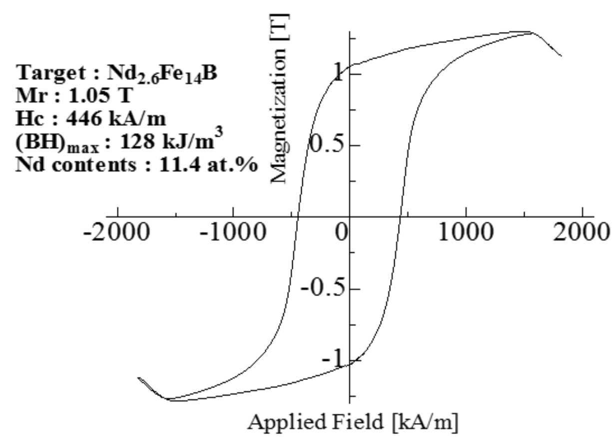


Fig. 9 M-H loop of a sample with the Nd content of 11.4 at. % seen in Fig. 8. The sample with $(BH)_{max}$ of 128 kJ/m^3 could be obtained by using a $Nd_{2.6}Fe_{14}B$ target ablated at LED≧10 J/cm^2.

B. *Improvement in magnetic properties of Nd-Fe-B thick-film magnets prepare by LED ≧ 10 J/cm²*

Although the $(BH)_{max}$ of a film prepared by LED ≧10 J/cm^2 reached up to approximately 110 kJ/m^3 as shown in Fig. 1, the value was inferior to those of isotropic RE (Nd or Pr)-Fe-B/α-Fe nano-composite ribbons with $(BH)_{max}$ above 130 kJ/m^3 [4][10] In order to improve the magnetic properties, an control of composition in the above-mentioned films was carried out by increasing the Nd amount of a Nd-Fe-B target as displayed in Fig. 8. It was clarified that an increase in Nd content in a film up to 11.5 at.% enabled us to enhance the value of $(BH)_{max}$ by approximately 20 kJ/m^3 compared with those of samples with the Nd contents range from 8 to 10 at.% (see Fig. 1). Furthermore, a M-H loop of a sample with the $(BH)_{max}$ of 128 kJ/$m^3$was shown in Fig. 9.

It was clarified that the improvement in $(BH)_{max}$ by controlling the film composition in PLD-fabricated nano-composite thick-film magnets prepared by LED ≧10 J/cm^2.

IV. Conclusion

In this study, we fabricated isotropic nano-composite thick-film magnets by using a PLD method with high laser energy density (LED) above 10 J/cm^2. The obtained results are summarized as follows;

(1) A nano-composite film with dispersed α-Fe + Nd-Fe-B structure could be formed and the average diameter of α-Fe grains was approximately 20 nm.

(2) Usage of a small spot size of a laser beam within an optimum deposition time enabled us to form the-above mentioned microstructure.

(3) The control of composition in the above-mentioned nano-composite film was effective to enhance the $(BH)_{max}$ value up to approximately 130 kJ/m^3.

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