Computer Simulation of Enhancement of Coerciviy in Nd-Fe-B/(Nd,Dy)-Fe-B Composite Magnets

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The coercivity H_c of $Nd_2Fe_{14}B$ magnets and $Nd_2Fe_{14}B/(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ composite magnets were calculated by computer simulation based on the micromagnetic theory under assumptions that $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains have magnetically deteriorated layers on their surfaces and diffusion of Dy from $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains to $Nd_2Fe_{14}B$ ones through the contacting boundaries recovers the magnetic anisotropy of the deteriorated layers of $Nd_2Fe_{14}B$ grains. H_c of $Nd_2Fe_{14}B/(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ composite magnets increased by the diffusion of Dy from $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains to $Nd_2Fe_{14}B$ ones and the resultant recovery of the anisotropy field of deteriorated layers of $Nd_2Fe_{14}B$ grains. The H_c vs fraction of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains curve were convex for the magnets with the degree of alignment between 0.94 and 0.99, which suggests that the above composite magnets have larger H_c values than the alloy-magnets with the same Dy content, and that we can save the consumption of Dy by using these composite magnets.

Keywords : (Nd,Dy)-Fe-B, coercivity, maicromagnetic simulation, magnetization reversal, diffusion of Dy, magnetically deteriorated layer

1. Introduction

Recently, Nd-Fe-B sintered magnets with excellent magnetic properties were widely used for electrical devices and equipments. For such applications, a magnet is needed to have a high coercivity value even at a high temperature, typically 200 °C. Although the substitution of a part of Nd for Dy is effective in increasing coercivity H_c, the resource of Dy is limited and it is necessary to suppress its consumption. As one of breakthroughs for this problem, it has been reported that H_c of a small Nd-Fe-B sintered magnet is increased by the grain interface reforming due to the diffusion of a heavy rare earth element such as Dy to the grain surface of $Nd_2Fe_{14}B$ [1, 2], which suggests that the importance of the surface of Nd-Fe-B grains. In this method, Dy was coated on surfaces of a magnet and was diffused to the surfaces of the grains of the magnet by annealing. Thus, the coated Dy played a role of Dy-source. The source of Dy is one of key points of this method and other sources have already reported [3, 4].

In this contribution, we assumed $Nd_2Fe_{14}B/(Nd,Dy)_2Fe_{14}B$

composite magnets and the diffusion of Dy from $(Nd,Dy)_2Fe_{14}B$ grains to $Nd_2Fe_{14}B$ ones. Then, we varied the fraction of $(Nd,Dy)_2Fe_{14}B$ grains, carried out a numerical simulation of magnetization reversal process in the composite magnets based on the micromagnetic theory, and clarified effects of the Dy diffusion on H_c of Nd-Fe-B/(Nd,Dy)-Fe-B composite magnets.

2. Simulation Method

2.1. Simulation model

The atomic ratio of Dy to (Nd+Dy) in $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains was set at 0.3 in this study, and the model magnet consisted of 8 cubic grains ($Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ ones). The number of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains was varied from 0 to 8. A grain, 48 nm in size, was divided into 4096 elements as shown in Fig. 1. The direction of the easy axis of $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains were distributed from that of the applied field by the Gaussian function [5] and the degree of deviation was evaluated by the average of $\cos\theta$, $<\cos\theta>$, where θ is the deviation angle of the easy axis of magnetization from that of an applied field. The $<\cos\theta>$ values of 1 and 0.5 correspond to perfectly aligned and isotropic magnets, respectively.
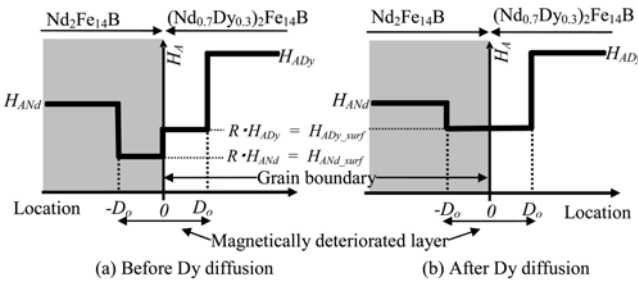
Figs. 2(a) and (b) shows the variations of the anisotropy

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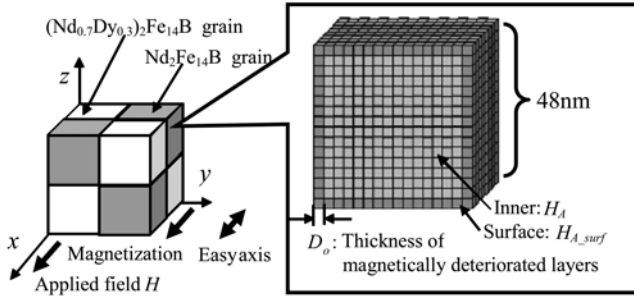
Fig. 1. Simulation model. Definitions of D_o, H_A, H_{A_surf} are indicated in the figure.

Fig. 2. Variations of anisotropy fields in the vicinity of boundary between $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains. Figs. (a) and (b) indicate the variations before and after Dy diffusion, respectively. In the figure, the subscripts, Nd and Dy indicate $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains, respectively, and R is the recovery rate. We assumed that diffusion of Dy recovers the anisotropy field of the deteriorated layers of $Nd_2Fe_{14}B$ grains up to that for $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ ones as show in (b).

field in the vicinity of $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grain boundaries. Both grains have the magnetically deteriorated layers, 3 nm in thickness, on their grain surfaces [6], and the anisotropy fields in the deteriorated layers were assumed to be $R \cdot H_A$, where H_A is the anisotropy field of the inner part of the corresponding grain. R is the recovery rate and varies between 0 and 1. The anisotropy fields of the inner parts of the $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains, H_{ANd} and H_{ADy} respectively,

were determined from the previous report for single crystals [7] as indicated in Table 1. The other simulation parameters are also shown in Table 1.

Furthermore, we assumed that annealing of a composite magnet causes the diffusion of Dy from $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains to $Nd_2Fe_{14}B$ ones through the contacting grain boundaries, and that the anisotropy field of a $Nd_2Fe_{14}B$ grain increases as indicated in Fig. 2(b), although that of a $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grain is kept constant.

2.2. Simulation method

We deduced the magnetic energy W stored in the model magnet mentioned above, considering the anisotropy, exchange, magnetostatic, and Zeeman energies. The stable direction of magnetization in each element was determined so as to minimize W for a given applied field. Details of our calculation method were reported elsewhere [8].

3. Simulation Results

3.1. $Nd_2Fe_{14}B$ Magnets

At first, we examined the effect of magnetically deteriorated layers on magnetic properties by varying R. Fig. 3 shows typical variations of a demagnetization curve for the model magnets composed of $Nd_2Fe_{14}B$ grains. The $<\cos\theta>$ value was set at 1. As seen from the figure, magnetization was reversed at once except the case of $R = 0$, and H_c increased with increasing R.

For the one-dimensional model with $<\cos\theta> = 1$, we can obtain the dependence of the nucleation field on R analytically by extending the calculation by Aharoni [9], and the nucleation field reduced by the anisotropy, h_n, is given by

$$h_n = \cos^2\varphi + R\sin^2\varphi, \tag{1}$$

$$\varphi = T\sqrt{-R + h_n}, \tag{2}$$

$$T = D_o\sqrt{H_A M_S/2}, \tag{3}$$

where D_o and H_A are the thickness of the deteriorated

Table 1 Simulation parameters.

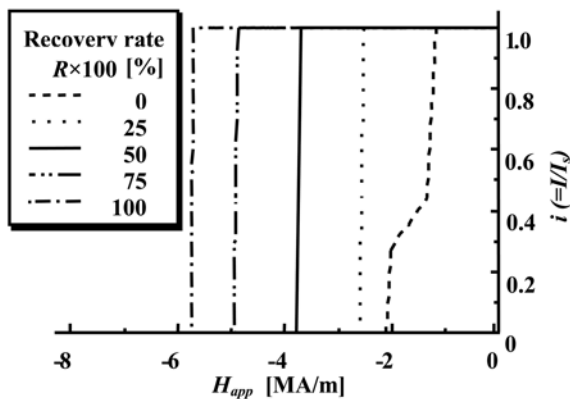| Parameters		Nd₂Fe₁₄B		$(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$
		Before diffusion	After diffusion	
Anisotropy field	- Inner -	5.60	5.60	6.54
H_A [MA/m]	- Surface -	2.80	3.27	3.27
Saturation magnetization	- Inner -	1.61	1.61	1.33
M_s [T]	- Surface -	1.61	1.33	1.33
Exchange stiffness constant A [J/m]		8.7×10^{-12}		
Grain size L [μm]		48		

Fig. 3. Demagnetization curves for model magnets composed of $Nd_2Fe_{14}B$ grains with different R values. The $<\cos\theta>$ value was set at 1.
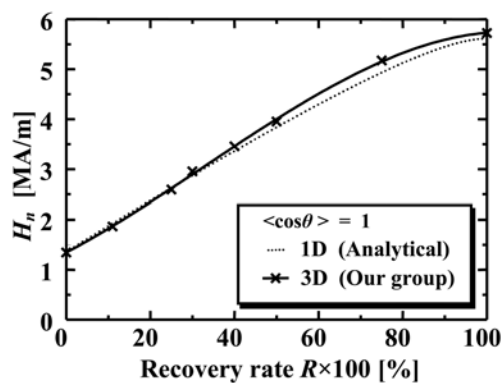

Fig. 4. Nucleation field H_n calculated by one- and three-dimensional models for perfectly aligned magnet ($<\cos\theta>$ = 1) as a function of the recovery rate R. The result for one-dimensional model was obtained by solving eqs. (1)-(3).

layer and the anisotropy field, respectively. The solution of eqs. (1)-(3) is shown in Fig. 4, together with that of our three-dimensional calculation in this study. As seen in the figure, both results are nearly same, suggesting the reliability of our simulation. The slight deference between analytical calculation and our result would be attributed to the difference in the models because our model is three dimensional and includes the effect of the magnetostatic interaction.

Demagnetization processes were simulated with varying $<\cos\theta>$ and R, and the obtained H_c is shown in Fig. 5 as a function of R. For the anisotropic magnets ($<\cos\theta>$ = 1, 0.96 and 0.92), H_c was improved significantly at R values around 0.5. On the other hand, H_c was not improved significantly for the isotropic magnets ($<\cos\theta>$ = 0.5), which suggests that the effect of magnetically deteriorated layers is not so significant for isotropy magnets. In order to clarify the origin of the little influence of the recovery for the isotropic magnets, we carried out the calculation
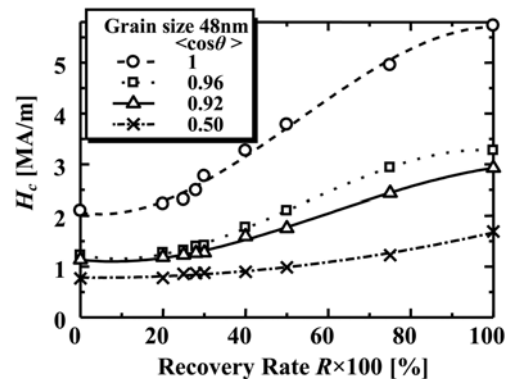

Fig. 5. Dependence of coercivity H_c of $Nd_2Fe_{14}B$ magnets on recovery rate R.

by omitting intergrain exchange interaction and found that H_c is improved even for isotropic magnets. This result strongly suggests that the averaging effect of anisotropy due to the intergrain exchange interaction suppresses the effect of the local variation in anisotropy due to the diffusion of Dy, and consequently the increase in coercivity.

3.2. $Nd_2Fe_{14}B/(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ composite magnets

Demagnetization process for $Nd_2Fe_{14}B/(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ composite magnets was simulated under the assumption of Dy diffusion. Fig. 6 shows the dependence of H_c on the fraction of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains with $<\cos\theta>$ as a parameter. The R value of $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains was set at 0.5 before diffusion, because the recovery of the magnetic anisotropy affects H_c significantly at $R >$ 0.5 as indicated in Section 3.1. Thus, the R value of a $Nd_2Fe_{14}B$ grain increases from 0.5 to 0.58 by diffusion, although that of a $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grain is kept constant. The dotted straight lines in the figure show the dependence of H_c for $(Nd_{1-X}Dy_X)_2Fe_{14}B$ magnets, which is described as alloy-magnets in this study, with the same Dy content with composite ones. Furthermore, the error bars indicate the simulation results for 7 model-magnets with different locations of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains and/ or different $\cos\theta$ values.

For the magnets with $<\cos\theta>$ = 0.98 and 0.97, H_c of the composite magnet increased remarkably, when the fraction of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grain became 0.25. Consequently, the H_c vs fraction of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ curves were convex, suggesting that the composite magnets have lager H_c values than the alloy-magnets with the same Dy content, and that we can save Dy consumption by using composite magnets. A similar tendency was obtained for the magnets with $<\cos\theta>$ = 0.99 and 0.94. The convex variation of H_c is reasonable because only 25% replacement of $Nd_2Fe_{14}B$ grains by $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ ones
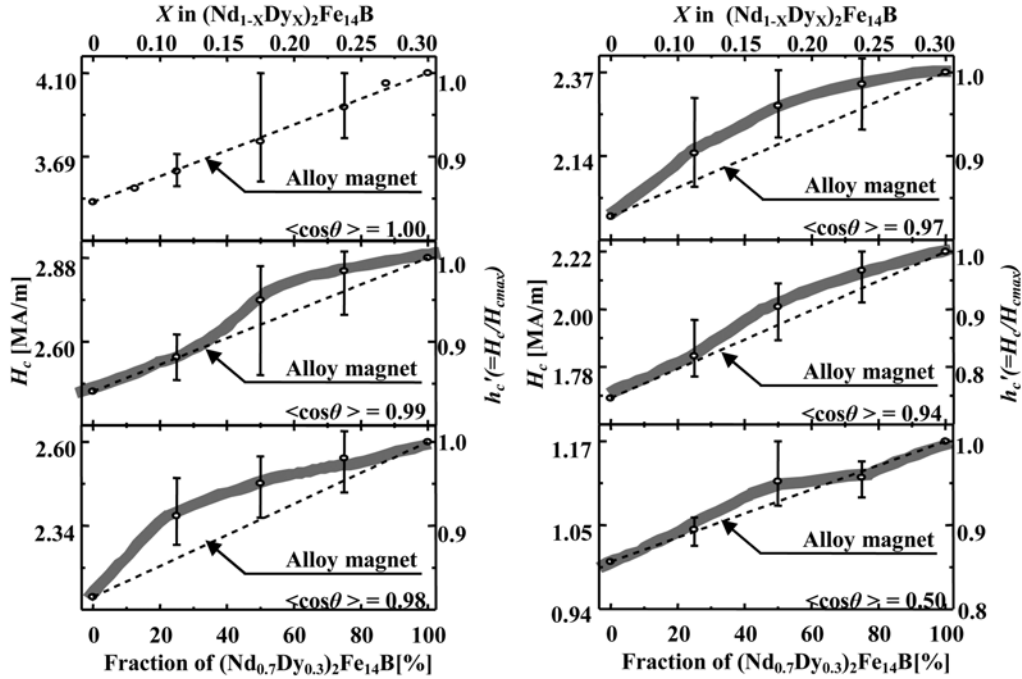
Fig. 6. Coercivity H_c of $Nd_2Fe_{14}B/(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ composite magnets as a function of fraction of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains. We assumed diffusion of Dy from $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains to $Nd_2Fe_{14}B$ ones and the resultant increase in R for $Nd_2Fe_{14}B$ grains. Details of the assumed variation of R are described in the text.

recovers the anisotropy field at 50% deteriorated layers.

On the other hand, H_c of the magnets with $<\cos\theta> = 1$ increased linearly with the fraction of $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$. This result is different from those for the magnets with $<\cos\theta> = 0.97$ and 0.98 shown in Fig. 6, which suggests that the magnetization reversal of perfectly aligned magnets ($<\cos\theta> = 1$) is strongly affected by unrecovered grain surfaces. In spite of the results for the magnets with $<\cos\theta> = 1$, it should be noted that the $<\cos\theta>$ values are less than 0.98 for practical magnets, and that the synthesis of composite magnets saves the Dy consumption for practical magnets.

4. Conclusions

The coercivity H_c of $Nd_2Fe_{14}B$ and $Nd_2Fe_{14}B/(Nd_{0.7}Dy_{0.3})_2$-$Fe_{14}B$ composite magnets were calculated by computer simulation based on the micromagnetic theory under assumptions that $Nd_2Fe_{14}B$ and $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains have magnetically deteriorated layers on their surfaces, and that diffusion of Dy from $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains to $Nd_2Fe_{14}B$ ones through the contacting boundaries recovers the magnetic anisotropy of the deteriorated layers of $Nd_2Fe_{14}B$ grains. Main results are summarized as follows.

The coercivity was improved by the recovery of the anisotropy field in magnetically deteriorated layers. Improve-ment in H_c was significant in highly anisotropic magnets, when the anisotropy field in the deteriorated layers exceeded the half of that of the inner parts.

For $Nd_2Fe_{14}B/(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ composite magnets, it was clarified that H_c increased by the diffusion of Dy from $(Nd_{0.7}Dy_{0.3})_2Fe_{14}B$ grains to $Nd_2Fe_{14}B$ ones and the resultant recovery of the anisotropy field of deteriorated layers of $Nd_2Fe_{14}B$ grains. The H_c vs fraction of $(Nd_{0.7}Dy_{0.3})_2$-$Fe_{14}B$ grains curve were convex for the magnets with $<\cos\theta> = 0.94$-0.99, which suggests that above composite magnets have larger H_c values than the alloy-magnets with the same Dy content, and that we can save Dy consumption by using these composite magnets.

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