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Dy gradient and coercivity in grain boundary diffusion processed Nd-Fe-B magnet

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Abstract: By controlling Dy vapor deposition process, the amount of Dy that diffused into the magnet was increased gradually from 0.1 wt.% to 0.3 wt.%. Compared with the original status, the coercivity increment was not proportional to the Dy diffusion amount. Subsequent H_{cj} and Dy content gradient data showed that slope of the 0.3 wt.% sample gradient was bigger than that of 0.1 wt.% one, and the gaps between outer flakes and inner flakes enlarged with the increasement of Dy diffusion amount. Although Dy mostly enriched in triple-junction regions in electron-probe microscope analysis (EPMA) images, the following Auger depth graph showed that Dy content was as high as 3.0 at.% in 1.5 mm deep center. It proved that Dy tended to get into the main phase rather than stayed in the grain boundary during the diffusion process, and over-diffusion of Dy in the main phase was unhelpful for the coercivity enhancement.

Keywords: Nd-Fe-B sintered magnet; grain boundary diffusion process; Dy content; coercivity; rare earths

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Nd-Fe-B permanent magnet, a core part of driving motor of newly developing electric vehicles (EVs) and hybrid vehicles (HEVs), has attracted great attention. However, high temperature environment within the engine requires extraordinarily high room-temperature coercivity for Nd-Fe-B magnet[1,2]. Recently, a newly developed process called grain boundary diffusion process (GBDP), provides a suitable solution for the problem mentioned above. With the help of various GBDPs, including HRE oxide/fluoride coating, HRE metal sputtering, or HRE metal vapor sorption method[3–5], Dy is able to diffuse along the grain boundary phase forming a Dy-rich shell around the $Nd_2Fe_{14}B$ grains[4], effectively improving the crystalline anisotropy of the grain boundary region. As a result, large enhancement of the coercivity and slight decrease of remanence are realized simultaneously.

However, despite all these efforts, in GBD process, HRE have to diffuse from the surface of the magnet and then permeate through the solid magnet bulk. This behaviour, inevitably, results in the limitation, i.e., the gradient distribution of HRE concentration at different depth from the magnet surface, which was already reported by several previous studies[6–11].

Obviously, as the most essential feature in GBDP magnets, Dy gradient distribution determines magnetic properties ultimately. Thus the investigation about the correlation between Dy distribution and intrinsic properties is necessary. In present work, the amount of Dy introduced into the magnet was increased gradially, aiming to find out internal relationship between microstructural gradient and magnetic performance in GBDP magnet.

1 Experimental

The starting material was a commercially available Nd-Fe-B sintered magnet (ATMAX35SH; AT&M Ltd. Co., Beijing, China), with a nominal composition of $Nd_{12.3}Fe_{bal}B_{6.5}Dy_{1.5}Al_{1.0}Cu_{0.4}Co_{0.8}$ (where the dimensions of the subscripts are at.%). All of the minor elements were added during melting process. The Dy diffusion treatment was implemented on the rectangular cubic samples with a dimension of 32 mm×12 mm×3 mm using a vapor deposition method, and the diffusion temperature ranged from 700 to 1000 ºC. Thereafter, an aging treatment was carried out at about 600 ºC with the protection of Ar atmosphere. The amounts of Dy diffused into the samples were controled by time duration of GBDP.

The magnetic properties of the block magnets were measured by a NIM-10000H BH tracer. Furthermore, the block samples were sliced equally into five flakes with the dimension of 12 mm×12 mm×0.6 mm in order to measure the distributions of Dy contents, and thereafter 2 mm×2 mm×0.6 mm samples were cut from the center of the flake for the subsequent Hcj distribution measurement (Fig. 1). Sliced flake magnetic properties were measured by Lake Shore 7410 VSM and the composition was analyzed by ICP (inductively coupled plasma mass

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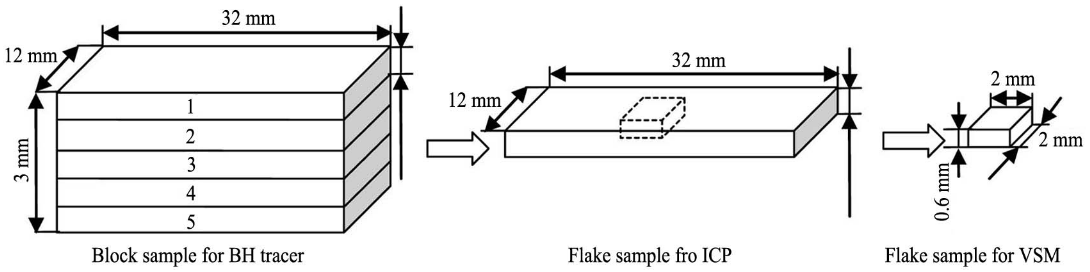

Fig. 1 Schematic illustration of samples by wire-cutting

spectrometer IRIS Intrepid). Microstructural observations were performed using an EPMA-1600. AES PHI-700 was used to take compositional profiles in the grain boundary region.

2 Results and discussion

Table 1 shows the sample mass changes induced by different time durations of Dy vapor deposition process, and the mass fraction of introduced Dy during the GBDP was calculated to be 0.09 wt.%, 0.19 wt.% and 0.29 wt.%, respectively. Hence, in the following part, we separate the samples into four categories, and they are named as “diffused 0.1 wt.%”, “diffused 0.2 wt.%”, “diffused 0.3 wt.%” and “ untreated”, where “untreated” represents the contrast sample without any treatment.

Fig. 2 and 3 show the demagnetization curves of the diffusion treated samples and the corresponding magnetic performance values respectively. As shown in Fig. 2, the coercivity-H_{cj} of the sample remarkably increases with the increase of Dy diffusion amount from 0.1 wt.% to 0.3 wt.%, but remanance-Br hardly decreases. Fig. 3 shows more detailed variations of coer civities and remanance upon the diffused Dy amount. Actually, the H_{cj} values of the Dy diffused samples reach as high as 2161 and 1922 kA/m with the diffusion of 0.3 wt.% and 0.1 wt.% Dy, respectively, and those are 449 and 210 kA/m higher than the H_{cj} of untreated sample. In the meanwhile, the decrease of Br upon the increase of Dy amount should not be neglected either, although decendent amplitude is quite small (no larger than 0.02 T).

Table 1 Mass change of samples treated by different time duration GBDPs

No.	$\text{Mass}_{\text{before}}$/g	$\text{Mass}_{\text{after}}$/g	Dy amount/wt.%
1	8.7626	8.7706	0.09
2	8.7295	8.7458	0.19
3	8.7532	8.7789	0.29

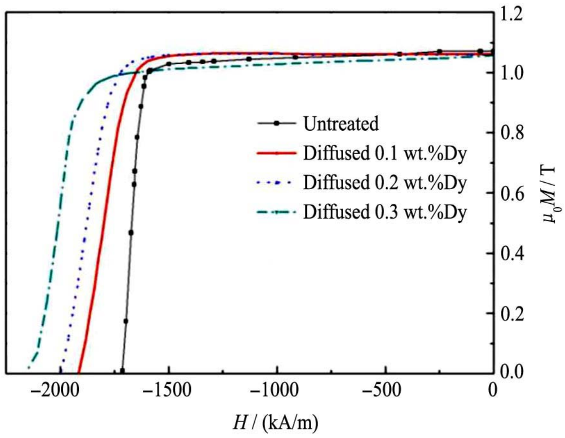

Fig. 2 Demagnetization curves of blocks diffused different amount of Dy

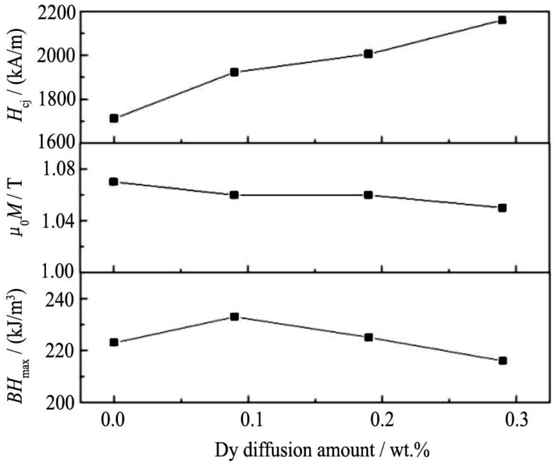

Fig. 3 Variation of magnetic performance upon different Dy diffusion amount

Previously, several reports have already dicovered that the most important trait of GBDP is remarkable enhancement of H_{cj} with little consumption of Dy, especially in report[5], H_{cj} increment rate per 1 wt.% rare earth diffusion amount was calculated. Thus, similar math was done in present work to compare the effeciency of Dy consumption after different amounts of Dy was diffused into the samples, as shown in Fig. 4. It is interesting to find out that, H_{cj} increment does not increase linearly with the increase of Dy diffusion amount and H_{cj} increment of 0.1 wt.% sample is almost half as much as 0.3 wt.% one. The other gragh in Fig.4, the variation of H_{cj} increment per 1 wt.% diffused Dy reveals this non-linear tendency more directly. The H_{cj} increment rate is as high as 2330 kA/m/wt.% for 0.1 wt.% sample while the value decreases to around 1550 kA/m/wt.% when Dy amount increases to 0.2 wt.% or 0.3 wt.%. That is to say, the very first 0.1 wt.% Dy diffused into the magnet increases H_{cj} much larger than the following 0.2 wt.% Dy thereafter.