Microstructure and coercivity of grain boundary diffusion processed Dy-free and Dy-containing Nd-Fe-B sintered magnets

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Achieving a high coercivity, $\mu_0 H_c$, above 3.0 T at room temperature is important for Nd-Fe-B permanent magnets for the application to traction motors of electric vehicles and wind turbines to avoid thermal demagnetization. Grain boundary diffusion (GBD) is promising to achieve the high coercivity without the significant loss of remanence that cannot be avoided with Dy alloying. In the GBD process, heavy rare-earth element (HRE) is diffused from the surface of the bulk along the grain boundaries, thereby forming HRE-rich shell on the surface of the $Nd_2Fe_{14}B$ main phase. This contributes to the coercivity increment. However, the coercivity cannot reach 3.0 T by the GBD process alone unless the initial sintered magnets are alloyed with Dy; a substantial amount of Dy must be alloyed in the base magnets in order to achieve the high coercivity of 3.0 T after the GBD process[1]. In the present study, we investigated the magnetic and microstructural characteristics of the GBD processed Dy-free and Dy-containing sintered magnets in order to understand the origin of the high coercivity of 3.0 T.

A GBD process was applied to two different sintered magnets with the chemical composition of $Nd_{14.3}Fe_{78.15}B_{6.0}Cu_{0.1}Al_{0.4}Co_{1.0}Ga_{0.05}$ and $Nd_{11.2}Dy_{3.1}Fe_{78.15}B_{6.0}Cu_{0.1}$ $Al_{0.4}Co_{1.0}Ga_{0.05}$ (at.%). The Dy-free and Dy-containing samples have the coercivity of 0.64 and 2.29 T before the GBD process. They were kept in Dy-vapor at 950 °C for 4 h for the GBD treatment followed by the post-diffusion annealing at 520 °C for 1 h. The Dy distributions in these magnets have been investigated via the use of SEM (Carl Zeiss Cross beam 1540EsB) and TEM (FEI Titan G2 80-200).

The coercivity increment, $\Delta\mu_0 H_c$, by the GBD treatment for the Dy-containing magnet was only 0.08 T, which was much smaller than that for the Dy-free magnet of 0.87 T due to the discontinuity of the Nd-rich GB phase in the Dy-containing magnet (Fig. 1). After the subsequent post-diffusion annealing, a substantial coercivity increase was observed, and the $\mu_0 H_c$ of the Dy-containing magnet reached 3.05 T while that of the Dy-free magnet saturates at 2.29 T. One reason for the 3T coercivity in the Dy-containing sample is the formation of the thick GB phase with high Nd content.

Interestingly, we found the formation of a “secondary Dy-rich shell” within the well-known primary Dy-rich shell[2]. The Dy element enriched in the Nd-rich GB phase during the GBD treatment is diffused into the main phase to form the secondary Dy-rich shell during the post-diffusion annealing. Such a secondary Dy-rich shell gave additional rise in the coercivity to 3T due to the increase in the Dy content at the GB phase/$(Nd,Dy)_2Fe_{14}B$ interface.

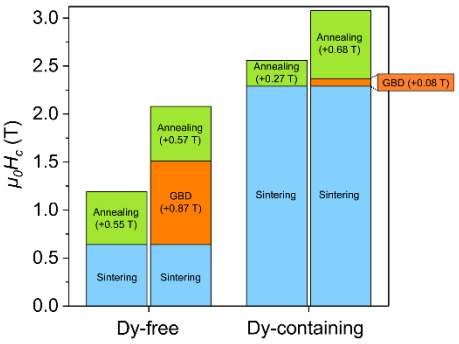

Fig. 1: $\Delta\mu_0 H_c$ of Dy-free and Dy-containing magnets with GBD process steps. For comparison, $\Delta\mu_0 H_c$ of magnets annealed without undergoing the GBD treatment are also displayed.

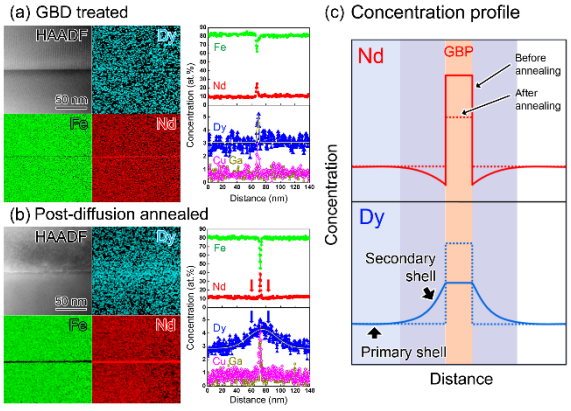

Figure 6: HAADF-STEM image and corresponding EDS elemental maps, and EDS line profiles across GB phase in (a) GBD treated and (b) post-diffusion annealed Dy-free magnet. (c) schematically shows the change in Nd and Dy concentration before and after the post diffusion annealing.

Reference
1) S.-E. Park et al., IEEE Trans. Magn, 47 (2011) 3259-3262
2) T.-H. Kim et al., Acta Mater. 175 (2019) 139-149