Temperature dependence of microstructure of Tb-rich shell in grain boundary diffusion processed Nd-Fe-B sintered magnets

Tae-Hoon Kim[a], T. T. Sasaki[a], T. Ohkubo[a], Y. Fujikawa[b], M. Miwa[b], Y. Enokido[b], and K. Hono[a]
[a] *Elements Strategy Initiative Center for Magnetic Materials, National Institute for Materials Science, Tsukuba-city, Ibaraki 305-0047, Japan*
[b] *TDK Corporation, Narita-city, Chiba 286-8588, Japan*

In Nd-Fe-B sintered magnets, the formation of heavy rare earth (HRE, Dy or Tb) enriched shell by the grain boundary diffusion process (GBDP) is essential to improve the coercivity while minimizing the HRE usage [1]. Due to higher anisotropy field of $Tb_2Fe_{14}B$ than $Dy_2Fe_{14}B$, the Tb-GBDP is more effective to enhance the coercivity of the magnets. During the GBDP of Tb at 970 ºC, the Tb-rich shell is formed by the diffusion induced grain boundary migration (DIGBM) [1]. The Tb concentration at the Nd-rich grain boundary (GB) region abruptly increases upon the GBD of Tb, and it provides a chemical driving force for the GB migration. Thereby, after the Tb-rich shell formation, the size of the main phase grain increases by the thickness of the shell [1]. This implies that the thickness of Tb-rich shell and the size of main phase grain can be controlled simultaneously by controlling the driving force for DIGBM. According to the simulation, the formation of thinner Tb-rich shell and smaller main phase grain are strongly required to further improve the coercivity of Tb-GBDP magnets [2], and we can expect that those microstructure is obtained by reducing the Tb-GBDP temperature. In this study, we investigated the change in the microstructure of Tb-rich shell as a function of the Tb-GBDP temperature.

The as-sintered Nd-Fe-B magnets were GBD processed using TbH_2 powder at 870, 920, and 970 ºC. The GBDP magnets were annealed at 520°C. Magnetic and microstructure characterizations were carried out using a BH-tracer, EDS, SEM, and TEM.

Fig. 1 shows the magnetic property changes as a function of the Tb-GBDP temperature. The coercivity of GBDP magnet increased from 1.9 to 2.1 T as the GBDP temperature decreased from 970 to 870 ºC. The remanence was not affected by the change of GBDP temperature. Fig. 2(a) shows the microstructure of 870 ºC and 970 ºC GBDP magnets at a depth of 100 μm. In the higher temperature GBDP magnet, the thickness of Tb-rich shell was thicker and the size of main phase grain was larger, compared to those in the lower temperature GBDP magnet. Notably, the average Tb concentration of the shell was higher in the lower temperature GBDP magnet, as shown in Fig. 2(a). Upon the GBD of Tb, the increment of Tb concentration at the Nd-rich GB region (*i.e.* driving force for DIGBM) was larger when the GBDP temperature was higher, thereby forming thicker Tb-rich shell and larger grain in the higher temperature GBDP magnet. Since the GB migrated while consuming the Tb during the Tb-rich shell formation, the average Tb concentration within the shell was higher when the thinner Tb-rich shell was formed at lower GBDP temperature, as shown in Fig. 2(a). Interestingly, the formation of Tb-rich shell was also observed at the center part of both the 870 ºC and 970 ºC GBDP magnets as shown in Fig. 2(b). Unlike near the magnet surface, the Tb-rich shell at the magnets center seems to be formed by the solid diffusion of Tb rather than the DIGBM. Nevertheless, as observed at 100 depth, thinner shell with higher Tb concentration was also formed at the center part of the lower temperature GBDP magnet, as shown in Fig. 2(b). Conclusively, the smaller grain size and the formation of thinner Tb-rich shell with higher Tb concentration are the main factor for higher coercivity of lower temperature GBDP magnet. The relationship between the GBDP temperature and microstructure of Tb-rich shell at various magnet depths will be discussed in detail.

References

[1] T. H. Kim *et al.*, to be submitted (2018).
[2] T. Oikawa *et al.*, AIP Advances, 6 (2016) 56006-1.

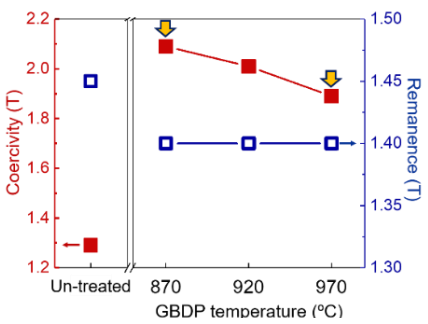

Figure 1. Magnetic property change of Tb-GBDP magnet as a function of GBDP temperature

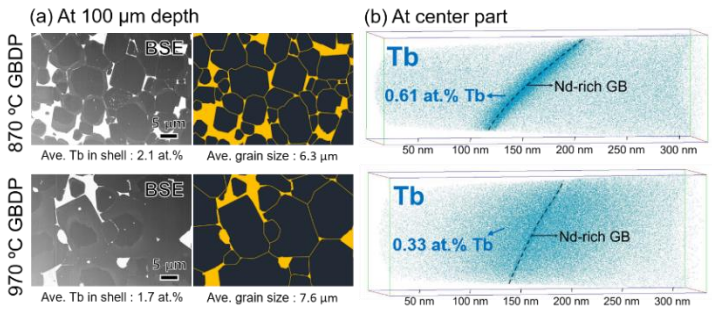

Figure 2. (a) BSE images and tracings of GB at 100 μm depth of Tb-GBDP magnets. (b) 3DAP atom maps for Tb at center part of Tb-GBDP magnets.