EFFECT OF DyMn ALLOY-POWDER ADDITION ON MICROSTRUCTURE AND MAGNETIC PROPERTIES OF NdFeB SINTERED MAGNETS

M.-W. LEE∗, D.R. DHAKAL∗, T.H. KIM∗∗, S.R. LEE∗∗, H.J. KIM∗∗∗, T.S. JANG∗♯

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1. Introduction

Magnetic properties, thermal properties and corrosion resistance of NdFeB sintered magnets can be improved by adding several elements. With excellent magnetic properties, NdFeB sintered magnets are used in the driving motor of a hybrid electric vehicle but usually unable to sustain at high temperature operation due to low Curie temperature and high temperature coefficient of the coercivity. This disadvantage can be overcome when their coercivity is enhanced by adding heavy rare-earth elements like Tb or Dy to NdFeB magnets by creating high anisotropy field [1-2]. However, the remanence reduction is always accompanied with the usage of these heavy rare-earth elements for the coercivity improvement. The antiferromagnetic coupling between heavy rare earth elements and Fe leads to reduce the saturation magnetization and the energy product \((BH)_{max}\). Moreover, limited resources and expensive price of Tb and Dy drive many researchers to seek a method to save heavy rare earths as much as possible, keeping high coercivity.

The addition of Mn on NdFeB magnet can be another method to enhance the coercivity, because it increases c-axis magneto-crystalline anisotropy by increasing c/a ratio of \(\text{Nd}_2\text{Fe}_{14}\text{B}\) lattices [3]. In addition, the wettability and continuity of Nd-rich phase can be enhanced by Mn addition because Mn decreases the melting point of Nd-rich phase [4]. However, large amount of Mn addition on the magnet reduces the saturation magnetization because non-magnetic Mn occupying \(J_2\) site loses ferromagnetic coupling with Fe atoms. In this work, we studied the effect of DyMn alloy powder addition on microstructure and magnetic properties of NdFeB sintered magnets and were able to improve the coercivity with minimum reduction of the remanence.

2. Experimental

Starting alloy with the composition of \(\text{Nd}_{14.7}\text{Fe}_{76.5}\text{B}_{6.0}\text{M}_{2.8}\) (M = Cu, Co, Al, Nb, at.%) was prepared by strip casting. The strips were HD treated and jet-milled to produce magnetic powder. The average particle size of the powder was about 5 µm. The jet-milled powder was wet-mixed with 1.2 wt% of DyMn alloy-powder. The solvent was removed by creating vacuum atmosphere. The mean particle size of DyMn alloy powder was about 4 µm. The mixed powder was aligned under the magnetic field of 2.2 T and pressed to form green compacts. The compacts were then sintered at 1070°C for 4 hours in vacuum, followed by a sequential annealing at 850, 530 and 500°C for 2 hours each in vacuum. Microstructural studies were carried out with a Scanning Electron Microscopy (SEM, Hitachi S-3000N). Phases were analyzed with Electron Probe Micro Analyzer (EPMA, SHIMADZU EPMA-1720). Magnetic properties were measured using a B-H loop tracer (Magnet Physik Permagraph C-300).

3. Results and discussion

Fig. 1 shows the magnetic properties of Dy-free magnets, DyMn-alloy powder mixed magnets (Dy 1.0 wt.%), and
the magnets containing 1.0 wt% Dy added by a conventional method. When Dy was added conventionally, the amount of coercivity increase was about 2.4 kOe, yielding 17 kOe. However, the coercivity was reached to 18.1 kOe, with the increase of 3.5 kOe, when small DyMn alloy-powder was mixed. Furthermore, the decrease of remanance by the mixing was very small (13.4 kG \rightarrow 13.1 kG). In order to figure out why the coercivity was improved more with DyMn alloy-powder mixing, without much decrease of the remanance, EPMA analysis on Dy and Mn distribution was carried out.

Fig. 1. Magnetic properties of Dy-free NdFeB sintered magnet, a DyMn alloy-powder mixed magnet (Dy 1.0 wt%), and a conventional Dy contained magnet (Dy 1.0 wt%)

As shown in the figure, however, most Mn was segregated at the Cu-rich triple junction phases (TJP) and the RE-rich phases. Typical composition of the Cu-rich TJP and the RE-rich phases was Nd_{40.2}O_{46.3}Cu_{3.1}Co_{5.0}Mn_{5.3} and Nd_{33.6}Dy_{1.6}O_{60.1}Mn_{4.6} (in at%), respectively. According to the previous results [9], the formation of the RE-rich phase deteriorates the coercivity of a NdFeB magnet because the surface energy of the RE-rich phase is very large. When Mn is segregated at the RE-rich phase, however, the surface energy seems to become lower because the melting point of RE-rich phase decreases due to Mn. This may help to reduce the lattice misfit between the RE-rich phases and the main phases. Furthermore, the wettability and the nonmagnetic property of the Cu-rich TJP would be improved by the Mn segregation also due to the low temperature melting behavior of Mn-contained nonmagnetic phases. As a result, the uniformity and continuity of the Nd-rich grain boundary phase (GBP) generated from the Cu-rich TJP would be improved too[10]. These effects are all beneficial to the improvement of the coercivity.

Fig. 2. EPMA mapping images of a sintered magnet mixed with 1.2 wt.% DyMn alloy-powder
Overall microstructures of a conventional Dy-contained sintered magnet (Dy 1.0 wt%) and a DyMn powder-mixed sintered magnet (Dy 1.0 wt%) are shown in Fig. 3(a). It is clearly seen that the magnetic grains are well isolated by continuously and uniformly developed Nd-rich grain boundary phase after DyMn alloy-powder mixing. As shown in Fig. 3(b), along with Cu, Mn is highly concentrated at Nd-rich grain boundary phase that developed from Cu and Mn enriched TJP. Cu-Mn binary phase diagram indicates that Cu and Mn are soluble to each other. Therefore, as Cu does, Mn would also penetrate through the Nd-rich GBP by a capillary action during sintering and improve the microstructural characteristics of Nd-rich GBP. Above results indicate that the coercivity of a NdFeB sintered magnet can be enhanced without much reduction of the remanence if the composition of DyMn alloy-powder source is kept to Dy-rich. Effect of the variation of Mn content in DyMn alloy-powder will be discussed in the near future.

4. Conclusions

The coercivity of a NdFeB sintered magnet mixed with small DyMn alloy-powder (1.2 wt%) increased from 14.6 to 18.1 kOe without much reduction of the remanence. Dy and Mn in the alloy-powder were decomposed during sintering and behaved differently. While Dy was mostly concentrated at the outer layer of the main grains forming a core-shell type structure, Mn was mostly concentrated at (Nd, Cu)-rich TJP and GBP improving the microstructural characteristics of Nd-rich GBP. The enhancement of the coercivity by the mixing of small DyMn alloy-powder was due to this duplicated effect of Dy and Mn although a minor reduction of the remanence arose from the substitution of some Fe with Mn was inevitable.

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