Microstructure and Properties of Die-Upset Nd-Fe-B/Dy₂O₃ Composite Magnets

Liyun Zheng¹,², Ke Zhang¹, Yanfeng Li¹, Xuchao Wang¹, Minggang Zhu¹, and Wei Li¹

¹Division of Functional Materials, Central Iron & Steel Research Institute, Beijing 100081, P.R. China
²School of Equipment Manufacturing, Hebei University of Engineering, Handan, 056038, P.R. China

Fabrication of rare-earth-based permanent magnets with both high electrical resistivity and excellent magnetic properties for the traction motor applications is highly challenging. In this paper, Nd-Fe-B/Dy₂O₃ composite magnets have been fabricated by hot-pressing and die-upset. The influence of Dy₂O₃ filler on the microstructures, electrical resistivity and magnetic properties of the Nd-Fe-B magnets have been investigated by scanning electron microscope, four-probe resistivity meter and hysteresis graph. The results showed that the electrical resistivities of the Nd-Fe-B/Dy₂O₃ composite magnets increased with the increase of Dy₂O₃ amount and reached 1270 μΩ cm when the Dy₂O₃ amount increased to 20 wt.%. Whereas the electrical resistivity of the nondoped Nd-Fe-B magnets is 230 μΩ cm. The cross-section SEM images of the composite magnets showed that the Dy₁₂O₃ phase formed laminated structure and some agglomerate phenomenon. The agglomerate areas became larger with the increase of Dy₂O₃ amount. The residual magnetic flux density (B_r) and maximum energy product of the Nd-Fe-B/Dy₂O₃ composite magnets decreased due to the nonmagnetic Dy₂O₃ filler. It is interesting to find that coercivity of the Nd-Fe-B/Dy₂O₃ composite magnets increased from 7.8 kOe to 9.6 kOe when the Dy₂O₃ amount increased from 10 wt.% to 20 wt.%. The reason may be that some of the dysprosium diffused into the Nd-Fe-B matrix and formed (Nd,Dy)Fe₁₄O₁₄ compounds. In the mean time, some of neodymium-rich phase diffused to Dy₂O₃ phase, replaced dysprosium and formed Nd₂O₃. This probably happened during the die-upset process at 850 °C.

Index Terms—Demagnetization, electrical resistance measurement, fabrication, magnetic properties.

I. INTRODUCTION

Nd-Fe-B magnets are now widely used in motors and generators due to their highest maximum energy product. However, the low Curie temperature of about 315 °C is the major practicable limitation for the traction motor applications of hybrid vehicles and electric vehicles. For these applications, the operating temperature of the motors increases to 200 °C due to eddy current produced in the magnets, which usually leads to thermal demagnetization of Nd-Fe-B alloys. To decrease the operating temperature, the NdFeB magnets with high eddy current resistance have been prepared by various processing techniques [1]–[3]. Electrically isolating Nd-Fe-B powders is an effective approach to fabricate magnets with high electrical resistance. Pan et al. [1] prepared isotropic NdₓFe₁₋ₓB magnets by coating the powders using a polymer mixture and the obtained magnets got a high resistivity of 940 Ω cm. But the maximum energy product (BH)ₓ max decreased to 4.3 MGOe. Furthermore, the operating temperature of the polymer bonded magnets is limited by the softening of the polymer.

By coating powders with fluoride, NdFₓ-coated hot-pressed Nd-Fe-B magnets with electrical resistivity of 1.4 m Ω cm were prepared and the loss in magnetism only reduced by 7/8 by using the fluoride-coated magnet [2]. Marinescu et al. [3] also fabricated the fluoride-added Pr-Fe-B die-upset magnets with electrical resistivity of 873, 580, and 115 μΩ cm by using 5 wt.% of CaF₂, NdF₃ and DyF₃, respectively. Furthermore, ferrite-coated SmₓFe₁₋ₓNₓ powders were fabricated and their compacting composite magnet got a high resistivity of about 4000 μΩ cm [4]. Gabay et al. [5] prepared SmCo₁₀/CaF₂ laminated composite magnets with electrical resistivity of 500–600 μΩ cm and maximum energy product of 6.5 MGOe by high energy milling and hot-pressing.

However, the magnetic properties of these composite magnets with high resistivity decreased to some extent due to the doping of insulated inorganic powders except Nd-Fe-B/NdF₃ magnets. It is believed that magnetic properties are not compatible with high resistivity because the Nd-rich grain boundary phase is necessary for a Nd-Fe-B magnet [6]. Dysprosium is another important element in the Nd-Fe-B magnets. DyF₃ was used to increase the eddy current resistance for Pr-Fe-B. But it is not as effective as CaF₂ to increase the resistivity of Pr-Fe-B magnets. Li et al. [7] investigated the distribution of Dy in high-coercivity (Nd,Dy)-Fe-B sintered magnet in details and found that Dy atoms are not uniformly distributed throughout the microstructure while some Dy oxides are present. There are some publications about the Dy₂O₃-added rare-earth permanent magnets [8]–[10], which did not investigate the electrical properties. In this paper, we investigated the influence of Dy₂O₃ on the microstructure, electrical resistivity and magnetic properties of the Nd-Fe-B magnet by hot pressing and die-upset. It is interesting to find that Nd-Fe-B/Dy₂O₃ composite magnet got a high resistivity of 1270 μΩ cm.

II. EXPERIMENTAL

NdₓFe₁₋ₓCo₄Ga₀₃B (wt.%) powders made by Magnequench were used as the starting materials and blended with 10 wt.%, 15 wt.% and 20 wt.% of Dy₂O₃ commercially available fine powders by grinding in a mortar. The mixtures were compacted in vacuum by hot pressing at 600 °C for 1 min. Hot-pressed isotropic Nd-Fe-B magnet can be deformed at 700 °C–900 °C. But the deformatability decreased due to the addition of Dy₂O₃. Thus, die-upsetting was carried out under...
an argon atmosphere at 850 °C for 100–120 s. The deformation rate was about 60%–70%.

The microstructure and microchemistry were examined by scanning electron microscopy (SEM) with a JEOL JSM-7001F with an energy dispersive x-ray detector. The magnetic properties were tested with a NIM-2000HF hysteresis graph meter after previous saturation with a pulse magnetizer at 100 kOe. The density of the composite specimen was measured by Archimedes’ principle. The resistivity was measured by the four probe electrical resistivity meter.

III. RESULTS AND DISCUSSION

Fig. 1 shows the microstructure of the magnets without and with Dy₂O₃. The magnet without Dy₂O₃ appeared layer-structure, which composes of NdₓFe₁₄B matrix phase (marked A) and Nd-rich grain boundary phase (small white particles like chains marked B). The composite magnets with different amount Dy₂O₃ fillers also had layer-structure, which compose of white Dy₂O₃ phase (large white particles like chains marked C), white Nd-rich grain boundary phase (small white particles like chains marked B), and NdₓFe₁₄B matrix phase (marked A). The Dy₂O₃ phase in the magnets also appeared layer-structure and some agglomerate phenomenon. The agglomerate became large with the increase of Dy₂O₃ amount.

Fig. 2 shows the backscattered electron SEM images of magnets with 15 and 20 wt.% Dy₂O₃ fillers. Table I gives the energy dispersive spectrum (EDS) results. It shows that the Dy₂O₃ filler parts were surrounded by the Nd-rich phase when the filler increased to 20 wt.%. This indicates that some Nd-rich phases melted, flowed to Dy₂O₃ and wrapped them during the hot deformation process. On the one hand, the Nd had a trend to diffuse to oxygen. On the other hand, hot deformation became difficult due to the higher amount of Dy₂O₃ filler and then the process time of hot deformation increased from 100 seconds to 120 seconds. Thus, the microstructure of Dy₂O₃ surrounded by Nd-rich phase formed. However, there is no this phenomenon in the samples with 10 wt.% and 15 wt.% fillers.

Fig. 3 shows XRD patterns of the magnets without filler and with different Dy₂O₃ content. All the composite specimens showed a [001] texture. The I_{006}/I_{105} values of the composite magnets with 10 wt.%, 15 and 20 wt.% of Dy₂O₃ were approximately 1.10, 1.54, and 1.66, where I_{006} and I_{105} are the integrated intensities of the (006) and (105) diffraction peaks. The I_{006}/I_{105} of NdₓFe₁₄B is 0.31 for an isotropic powder specimen [11]. The I_{006}/I_{105} values of the composites magnets increased with the increase of the amount of Dy₂O₃. This indicates that the deformation degree increased with the increase of the Dy₂O₃ content and the fully deformation of the
composites magnets with higher amount filler due to its longer deformation time.

The electrical resistivities of the composite magnets are 420, 830, and 1270 $\mu\Omega$ cm when the amount of Dy$_2$O$_3$ filler are 10, 15, and 20 wt.%, respectively. The electrical resistivity of the magnet without filler is 230 $\mu\Omega$ cm. The resistivities of the composite magnets with 10, 15, and 20 wt.% Dy$_2$O$_3$ increased approximately 83%, 261%, and 453% times higher than that of the magnet without filler. The resistivity of the composite magnet with 20 wt.% Dy$_2$O$_3$ filler almost reached that resistivity of the fluoride, NdF$_x$-coated hot-pressed Nd-Fe-B magnets [2]. It can be concluded that the addition of Dy$_2$O$_3$ is another effective way to improve the electrical resistivity of Nd-Fe-B magnets.

However, their magnetic properties decreased much more than that of the fluoride, NdF$_x$-coated hot-pressed Nd-Fe-B magnets due to the large amount of nonmagnetic fillers [2].

Fig. 4 shows demagnetization curves of the composite magnets without filler and with 10, 15, and 20 wt.% of Dy$_2$O$_3$. The maximum energy products (BH)$_{max}$ of the composite magnets without filler and with 10, 15, and 20 wt.% of Dy$_2$O$_3$ are 49.9, 29.6, 28.1, and 20.1 MGOe, respectively. Their coercivities and residual magnetic flux densities ($B_r$) are 7.8, 8.49, 9.64, and 11.6, 10.91, and 9.53 kGs when the fillers are 10, 15, and 20 wt.%, which are lower than that of the magnet without filler. Compare with the magnet without filler, the coercivity and the maximum energy products of the Nd-Fe-B/Dy$_2$O$_3$ composite magnets with Dy$_2$O$_3$ decreased 28% and 59%, respectively. The reason that magnetic properties of the Nd-Fe-B/Dy$_2$O$_3$ composite magnets with Dy$_2$O$_3$ decreased is that the deformation of the hot-pressed composite magnet with Dy$_2$O$_3$ fillers became difficult and their formed texture and anisotropy were influenced. It is interesting to note that the coercivities increased from 7.8 kOe to 9.64 kOe when the fillers increased from 10 to 20 wt.%. The reason may be that some of the dysprosium diffused into the Nd-Fe-B matrix and formed (Nd,Dy)$_{14}$Fe$_{14}$B compounds. In the mean time, some of Nd-rich phase diffused
to Dy$_2$O$_3$ phase, replaced dysprosium and formed Nd$_2$O$_3$. This probably happened during the die-upset process at 850 °C.

IV. CONCLUSION

Nd-Fe-B/Dy$_2$O$_3$ composite magnets have been fabricated by hot-pressing and die-upset. The electrical resistivities of the composite magnets increased from 230 $\mu$Ω cm to 420, 830 and 1270 $\mu$Ω cm when the Dy$_2$O$_3$ amount are 10, 15, and 20 wt.%. The cross-section of the composite magnets formed laminated-structure of Dy$_2$O$_3$ and some agglomerate phenomenon. The agglomerate areas became larger with the increase of Dy$_2$O$_3$ amounts. The $B_r$ and $(BH)_{max}$ of the Dy$_2$O$_3$-added magnets were decreased due to the nonmagnetic Dy$_2$O$_3$ filler. It is interesting to find that the coercivities of the composite magnets increased from 7.8 to 9.64 kOe with the Dy$_2$O$_3$ amount increased from 10 to 20 wt.%.

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REFERENCES

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