# AN ANALYSIS OF THE MAGNETIZATION BEHAVIOR AND TEMPERATURE DEPENDENCE OF COERCIVITY IN HOT DEFORMED Nd<sub>2</sub>Fe<sub>14</sub>B MAGNETS WITH DIFFERENT DEFORMATION DEGREES

### Xu Tang\*, Renjie Chen, Xin Tang, Don Lee, Aru Yan\*

 Key Laboratory of Magnetic Materials and Devices, Ningbo Institute of Material Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, People's Republic of China
 Zhejiang Province Key Laboratory of Magnetic Materials and Application Technology, Ningbo Institute of Material Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201,

People's Republic of China

### Abstract

Hot deformation magnets with different height reductions are prepared following hot press process. The coercivity of the magnets decreases from 19.5kOe to 15.5kOe while the remanence increases from 8.5kGs to 13.5kGs. Minor loops of the hot pressed and deformation magnets are both measured to analyze the magnetization behaviors. The saturation magnetization vs. the maximum applied field of the hot pressed and deformation magnets are also carried out. The microstructures of these magnets are analyzed through scanning electron microscope (SEM). It can be obtained that the shape of the grains in these magnets changes into platelike when the height reductions get higher. The temperature dependences of the coercivity of the magnets are also obtained from the linear relationship between Hc/Ms and HA/Ms. In this paper, it is believed that the varieties of the coercivity is strongly dependant on the shape of the grains and the grain boundary structures.

# Keywords

coercivity, nanostructural parameter, coercivity mechanism, hot deformation magnets;

### Introduction

High performance NdFeB magnets have many commercial and military applications nowadays. Green energy technologies, such as the wind turbine and hybrid power automobile, are strongly demanded to exploit under the background of the energy resource crisis. The maximum energy product of the anisotropic nanocrystalline NdFeB magnets almost approaches the theoretical value reported by Lee.[1]. However, the coercivity of the magnets is only about 20~30% of the anisotropy field at room

temperature. The relatively low coercivity has been the main obstacle for more applications although it can be compensated by substituting the heavy rare earth Dy or Tb for Nd because of the much higher magnetocrystalline anisotropy[2, 3]. In order to enhance the coercivity and make use of the heavy rare earth more sufficiently, it is important to understand the coercivity mechanism in NdFeB permanent magnets. There are reports that pinning mechanism is the primary coercivity mechanism in hot deformation magnets[4]. Furthermore, the coercivity of the hot deformed magnets decreases by increasing the deformation degrees[5]. It is well known that the coercivity of the magnets is strongly influenced by the nanoctructural parameters described by Brown's equations [6-8]. The shape of grains and surface-to-volume ratio is changed along with the deformation degrees.

In nanocrystalline SmFeSiC ribbons, the magnetization behaviors can be influenced by intergrains exchange coupling (IGEC). The mechanism of magnetization reversal in such ribbons is mainly controlled by the inhomogeneous pinning of nucleated type reported by Hong-Wei Zhang[9]. However, it is not clear that whether or not IGEC still exists when the shape of the grains changes from equiaxed to plate-like.

In this work, we report the magnetization behaviors of the isotropic and anisotropic magnets with a deformation degree of 70%. The temperature dependences of coercivity of the magnets with different hot deformation degrees are discussed.

# Experiment

Magnets with different deformation degrees were prepared by hot deformation process at 1125K following hot press process at 945K under vacuum. The samples were cut into  $1 \times 1 \times 3$ mm for magnetic property measurement. The easy axis was the longitudinal direction. The magnetic properties, recoil curves and temperature-dependent coercivity of the magnets hot deformed at different degrees were obtained using a superconducting quantum interference device (SQUID) magnetometer with the maximum field of 40 kOe applying along the longitudinal direction of the samples. Plots of the saturation magnetization vs. the maximum applied field of the hot pressed and deformation magnets are obtained from the recoil curves. The temperature dependence of coercivity was measured at temperatures ranging from 300K to 500K.  $H_c / M_s$  vs.  $H_A / M_s$  plot for the hot pressed magnet and the die-upset magnets with different deformation degrees was achieved.

**Results and discussion** 



Figure 1. Saturation magnetization vs. the maximum applied field of the hot pressed and deformation magnets, the saturation magnetization obtained from the minor loop. + represents the values obtained in the first quadrant, and – those in the second quadrant.

Plots of the saturation magnetization versus the maximum applied field of the hot pressed and deformation magnets are shown in Fig. 1(a) and (b), respectively. The saturation magnetization Ms is obtained from the minor loop. The signal "+" represents the values which are obtained in the first quadrant and - those in the second quadrant. Both of the magnets are thermally demagnetized before measurement. The saturation magnetization at positive magnetization direction is lower than negative one for both of the magnets when the applied field is smaller than coercivity. The difference between positive and negative magnetizing directions may be caused by inhomogeneous domain wall pinning because of the inhomogeneous microstructure. The coercivity of the hot pressed magnets seems to be controlled by nucleation while the inhomogeneous pinning cannot be neglected at the same time. An eclectic explanation is that both pinning and nucleation determine the magnetization reversal. The same result also can be obtained for the hot deformation magnet at positive magnetization direction. However, a mechanism similar to pinning is observed at negative magnetization direction shown in Fig. 2(b). The domain walls in the thermally demagnetized magnets may exist at grain boundary and in grains where the domain wall energy is relatively low. Normally, the domain walls move easily inside grains. The domain walls are easily impelled to move from initial position to another one where the domain walls are pinned, while a negative field is applied, it is hard to move the domain walls away. The saturation magnetizations between positive and negative applied directions are equal while the external field exceeds the coercivity of the magnets. It means that most of the magnetic moments are

along with the applied direction through domain wall motion and de-pinning.



Figure 2. The SEM of the magnets prepared by hot deformation with different deformation degrees.

The microstructures of the magnets with different deformation degrees are analyzed through SEM. The deformation degrees are 43%, 57% and 70% for the magnets which are shown in Fig. 2(a), (b) and (c), respectively. There have been reports that the deformation degree of 70% is most suitable for preparing high performance anisotropic nanocrystalline magnets[5]. In this work, fine plate-like grains are also obtained which is shown in Fig. 2(c) through hot deformation with deformation degree of 70%. Besides, plate-like grains are observed in all of the magnets, in the meantime, some equiaxed grains are also existed in Fig. 1(a) and (b). The C-axes of the grains shown in Fig. 2(a) and (b) which is perpendicular to the base plane of the grains are not perfectly arranged at the same direction. The grain size increases along the base plane of grains when the deformation degree increases. However, there is little change of the grain size along short axes although the deformation varies. It is generally known that the remanence enhances while the coercivity largely decreases by increasing the deformation degree.

Magnetic properties of the magnets are measured and compared with different deformation degrees. As it is revealed in most works, the coercivity decreases from 19.5kOe to 15.5kOe while the remanence enhances from 8.5kGs to 13.4kGs by increasing the height reduction which is shown in Fig. 3.



Figure 3 The demagnetization curves of the magnets prepared by hot press and deformation process with different deformation

#### degrees

In order to describe the effect of the microstructure on the magnetic properties, coercivity as a function of temperature is measured for the magnets with different deformation degrees. It is obviously that different microstructures can be resulted from different height reductions. The temperature dependence of coercivity can be described by Brown's equation with appropriate modification.

$$H_{ci}(T) = \alpha_{\psi} \alpha_{\kappa} \alpha_{ex} H_A(T) - N_{eff} M_s(T)$$
<sup>(1)</sup>

Here  $H_A$  and  $M_s$  are the temperature dependence of magnetocrystalline anisotropy field and saturation magnetization of the hard 2:14:1 phase. The nanostructure parameters  $\alpha_{\psi}$ ,  $\alpha_{K}$ ,  $\alpha_{ex}$  and  $N_{eff}$  are related to different nanostructural effects.  $\alpha_{\psi}$  is related to the grain orientation distribution in the magnets with the value 0.5 for hot pressed magnets which there is no texture and 1.0 for perfectly aligned samples. The reduction of anisotropy at inner interfaces such as grain boundaries and crystal imperfections is taken account which is represented by nanostructural parameter  $\alpha_{K}$ . It can be increased by increasing the crystalline quality of the hard magnetic phases.  $N_{eff}$  is an effective demagnetization factor which is introduced to describe the internal stray field. The more angular edges exist in magnets, the larger the value of  $N_{eff}$  is. A smaller value of  $N_{eff}$  could be obtained by the less polyhedral grains and the smoothing effect of the stronger exchange coupling at the grain boundaries[5]. It indicates a reduction of the stray fields at the edges and corners on the grains.  $\alpha_{ex}$ which is always less than 1 describes the effect of exchange coupling between neighboring grains on the coercivity of the nanostructural magnets. The existence of the exchange coupling would lower the coercivity. Stronger exchange coupling will result in smaller value of  $\alpha_{ex}$ .

In this work, the modified Brown's equation analysis was carried out for hot pressed magnet and die-upset magnets which are deformed with deformation degrees of 57% and 70%. The temperature is arranged from 300K to 500K.  $H_c/M_s$  as a function of  $H_A/M_s$  is shown in Fig. 4.



Figure 4.  $H_c / M_s$  vs.  $H_A / M_s$  plot for the hot pressed magnet and the die-upset magnets with different deformation

#### degrees

The values of HA were taken from literature[10]. It is generally known that the C-axes of the grains in hot pressed magnets are distributed randomly.  $\alpha_{\psi}$  in hot pressed magnet is considered to be 0.5. The value of  $\alpha_{\psi}$ ,  $\alpha_{K}$  and  $\alpha_{ex}$  cannot be obtained separately from the temperature-dependent coercivity. However, a fitted parameters  $C = \alpha_{\psi} \times \alpha_{K} \times \alpha_{ex}$  represents the slope of the plots is given for later discussion. Preferable linear relationship is obtained in this work. It can be obviously seen that the slopes of the plots are almost the same. Even though the value of  $\alpha_{\psi}$  could not be obtained exactly,  $\alpha_{\psi}$  increases as the C-axes of the plate-like grains tend to be alignment through higher deformation degrees. The grain sizes grow up which is shown in Fig. 2 when the deformation degrees are higher, so the composition and width of the grain boundaries should have been inhomogeneity and drastically changed. It cannot be achieved that which one between  $\alpha_{K}$  and  $\alpha_{ex}$  is varied more. The crystal imperfections and reduction of anisotropy near the grain surfaces will result in a smaller value of  $\alpha_{K} \cdot \alpha_{ex}$  may be smaller because of the weaker exchange coupling between neighboring grains while the grain sizes increase. The value of  $N_{eff}$  for the magnets gets bigger as the deformation degrees become higher. The bigger value of  $N_{eff}$  suggests higher internal demagnetization fields ,which results from more angular grains with more sharp edges, when the shape of the grains varies from almost spherical to plate-like by increasing the hot deformation degree. The nanostructural parameter  $N_{eff}$  would be a major factor that will decrease the coercivity of the hot deformed magnets.

# Conclusion

Hot pressed magnet and hot deformation magnets with different degrees are prepared through hot press and deformation process. Saturation magnetization vs. the maximum applied field of the hot pressed and deformation magnets is obtained. Both nucleated and pinned types are analyzed in the hot pressed magnet and hot deformed magnet with a deformation value of 70%. SEM observation shows that the average size of the grains in these magnets increases and becomes plate-like by increasing the hot deformation degrees.  $H_c / M_s$  vs.  $H_A / M_s$  plots for the hot pressed and die-upset magnets with different deformation degrees are carried out. The shape of the grains could drastically affect the coercivity of the magnets. The shape of plate-like would be the primary factor which results in inferior coercivity in hot deformed magnets.

# Acknowledgment

This work was supported in part by National Natural Science Foundation (No. 60901047), the Minist ry of Science and Technology International Cooperation Project (No. 2010DFB53770), the China Postd octoral Science Foundation (No. 2012M520943), the State Key Program of National Natural Science F oundation of China (Grant No. 2010AA03A401) and National key technologies R&D Program of Chin a (No. 2012BAE01B03).

### References

- S. Liu et al., "Enhancing magnetic properties of bulk anisotropic Nd-Fe-B/alpha-Fe composite magnets by applying powder coating technologies," *IEEE Transactions on Magnetics*, 42(2006), 2912-2914
- [2] M. H. Ghandehari, "Reactivity of Dy<sub>2</sub>O<sub>3</sub> and Tb<sub>4</sub>O<sub>7</sub> with Nd<sub>15</sub>Fe<sub>77</sub>B<sub>8</sub> Powder and the Coercivity of the Sintered Magnets," Applied Physics Letters, 48(1986), 548-550.
- C. H. De Groot et al., "Two-powder Nd<sub>2</sub>Fe<sub>14</sub>B magnets with DyGa addition," Journal of Applied Physics, 83(1998), 388-393.
- [4] F. E. Pinkerton and C. D. Fuerst, "Coercivity of Die Upset Nd-Fe-B Magnets a Strong Pinning Model," Journal of Magnetism and Magnetic Materials, 89(1990), 139-142.

- [5] C. D. Fuerst and E. G. Brewer, "High-Remanence Rapidly Solidified Nd-Fe-B Die-Upset Magnets," Journal of Applied Physics, 73(1993), 5751-5756.
- [6] D. Goll, M. Seeger, and H. Kronmuller, "Magnetic and microstructural properties of nanocrystalline exchange coupled PrFeB permanent magnets," Journal of Magnetism and Magnetic Materials, 185(1998), 49-60.
- J.Bauer et al., "Nanocrystalline FeNdB permanent magnets with enhanced remanence," Journal of Applied Physics, 80(1996), 1667-1673.
- [8] B. Z. Cui et al., "Enhancement of exchange coupling and hard magnetic properties in nanocomposites by magnetic annealing," Acta Materialia, 53(2005), 4155-4161.
- [9] H. W. Zhang, S. Y. Zhang, and B. G. Shen, "Magnetic viscosity and coercivity analysis in nanocrystalline Sm<sub>2</sub>Fe<sub>15</sub>Si<sub>2</sub>C ribbons," Physical Review B, 62(2000), 8642-8645.
- [10] S. Hirosawa et al., "Magnetization and magnetic-anisotropy of R<sub>2</sub>FE<sub>14</sub>B measured on single-crystals," Journal of Applied Physics, 59(1986), 873-879.