Effect of Weak Magnetic Intergranular Phase on the Coercivity in the HDDR Nd-Fe-B Magnet

M. LIU, G. B. HAN, R. W. GAO

School of Physics, Shandong University, Jinan, China.
Email: hangb@sdu.edu.cn

Received July 2nd, 2009; revised August 11th, 2009; accepted August 19th, 2009.

ABSTRACT
Assuming that intergranular phase (IP) existing between adjacent grains is a weak magnetic phase, we study the effect of IP on the coercivity in the HDDR Nd-Fe-B magnet. The results indicate that the coercivity increases with the increasing IP’s thickness \( d \), but decreases with increasing its anisotropy constant \( K_1(0) \). When the structure defect thickness \( r_0 = 6 \) nm, \( d = 1 \) nm and \( K_1(0) = 0.15K_1 \) (\( K_1 \) is the normal anisotropy constant in the inner part of a grain), our calculated coercivity is in agreement with available experimental data.

Keywords: HDDR Nd-Fe-B Magnet, Intergranular Phase, Coercivity

1. Introduction
The HDDR powder particles, prepared by the HDDR (hydrogenation, decomposition, desorption, and recombination) process, consist of fine \( \text{Nd}_2\text{Fe}_{14}\text{B} \) crystalline grains with diameters ranging from 0.2 to 0.3 \( \mu \)m, which is close to the single domain size of \( \text{Nd}_2\text{Fe}_{14}\text{B} \) phase [1]. Such unique grain microstructure of HDDR magnet is different from not only the grain microstructure of sintered magnet, but also that of nanocrystalline magnet. Generally, the sintered magnet consists of the \( \text{Nd}_2\text{Fe}_{14}\text{B} \) crystalline grains of 5-10 \( \mu \)m in diameter, and nonmagnetic Nd-rich boundary phases [2] which interrupts the intergrain exchange coupling interaction. Thus, the grain-boundary anisotropy (GBA) of the sintered magnet is mainly affected by the grain-boundary structure defect (GBSD). The nanocrystalline magnet is composed of the directly contacted magnetic grains of a few tens of nanometers [3], and its GBA is principally influenced by the intergrain exchange coupling interaction (IECI). However, for the HDDR magnet, its GBA may be simultaneously influenced by GBSD and IECI [4], owing to the unique grain microstructure. Some investigators considered that the adjacent grains directly contacted with each other in the same HDDR powder particle [5,6,7]. However, Nakayama et al [8] observed experimentally that a thin grain-boundary layer with the thickness of 1 nm exists between adjacent HDDR grains. Theoretically, the effect of intergranular phase (IP) on the coercivity is unclear. Thus, this paper tries to theoretically study the effect of intergranular phase on the coercivity in HDDR Nd-Fe-B magnet.

The component, structure and character of intergranular phase sensitively depend on the alloy’s composition and processing technique. The intergranular phase is the crystalline phase with \( \text{Nd}_2\text{Fe}_{14}\text{B} \)-like structure reported by Reference [9]. Reference [10] pointed out that the \( \text{Nd}_6\text{Fe}_{13}\text{Al}_1 \) phase was identified as an intergranular phase. Thus, the intergranular phase is still magnetic phase. Here, assuming that the IP existing between adjacent grains is a weak magnetic phase, and using cubic-grain anisotropy model, we study the effect of IP on the coercivity of the HDDR Nd-Fe-B magnet. The results indicate that the coercivity increases with the increasing IP’s thickness \( d \), but decreases with increasing its anisotropy constant \( K_1(0) \). Such conclusion could provide a theoretical reference for preparing high coercivity HDDR Nd-Fe-B magnet.

2. Anisotropy Model
Reference [4] pointed out that the GBA is simultaneously influenced by the GBSD and IECI in the HDDR magnet, and proposed a structure model of a cubic grain with edge of \( D \) (where the GBSD’s thickness is \( r_0 \) and the IECI’s length is \( \text{lex} \)). Here, we assume that the IP is a weak magnetic phase, and distributes homogeneously between grains. Because of the very small size of IP, we presume that half of the thickness, \( d/2 \), is shorter than both \( \text{lex}/2 \) and \( r_0 \) (as shown in Figure 1a where \( r_0>\text{lex}/2 \) is supposed). IP weakens the IECI, leading to the IECI’s length reduce from \( \text{lex}/2 \) to \( (\text{lex}–d)/2 \). Based on different
ranges influenced by the GBSD and IECI, a grain is divided into three parts in the case of $D/2 > r_0 > \text{lex}/2$. For convenience, the center of IP is chosen as the coordinate origin of $r$. For $d/2 < r < \text{lex}/2$, the GBA is simultaneously affected by the GBSD and IECI. For $\text{lex}/2 < r < r_0$, it is influenced by the GBSD alone. While $r > r_0$, the GBA isn’t influenced by the GBSD or IECI, and is still the common anisotropy constant $K_1$ in the inner part of the grain. The grain-boundary anisotropy $K'_1(r)$ was described by different formulae for $r_0 \leq \text{lex}/2$ and $r_0 > \text{lex}/2$ in Reference [4]. Here, we assume that $K_1(0)$ is a constant in the IP region. Due to the continuous variation of $K'_1(r)$, its expression can be rewritten as Equations (1) and (2). Figure 1b shows the variation of $K'_1(r)$ in the case of $D/2 > r_0 > \text{lex}/2$. It can be seen that $K'_1(r)$ continuously decreases from $K_1$ in the inner part of a grain to $K_1(0)$ in the IP region.

when $r_0 > \text{lex}/2$,

$$
K'_1(r) = \begin{cases} 
K_1(0), & 0 \leq r \leq \frac{d}{2} \\
K_1 - \Delta K_1 \left(1 - \frac{2(r - \frac{d}{2})^2}{\left(r_0 - \frac{d}{2}\right)(\text{lex} - d)}\right)^{\frac{3}{2}}, & \frac{d}{2} < r < r_0 \\
K_1 - \Delta K_1 \left(1 - \frac{(r - \frac{d}{2})^2}{(\text{lex} - d)}\right)^{\frac{3}{2}}, & r_0 \leq r \leq \frac{\text{lex}}{2} \end{cases}
$$

Figure 1. (a) Sketch of a grain divided into three parts due to different ranges influenced by GBSD and IECI in the case of $D/2 > r_0 > \text{lex}/2$; (b) Variation sketch of grain-boundary anisotropy

3. Coercivity of the HDDR Nd-Fe-B Magnet

The demagnetization process and coercivity mechanism of the HDDR Nd-Fe-B magnet were studied by Reference [4], where the IP didn’t exist, and it was concluded that both the demagnetization nucleation and pinning of domain wall displacement between grains might occur at the grain boundary. If IP exists, it might become the pinning center of the domain wall displacement [11]. When the coercivity of magnet is determined by the irreversible domain wall displacement in the IP region, it can be expressed by [12],

$$
H_c = \frac{2K_1\pi r_0}{3\sqrt{3}M_s\delta_B} \left(\frac{A}{A'} \frac{K'_1}{K_1}\right) - N_{\text{eff}}M_s
$$

where $A$, $A'$ and $K_1, K'_1$ denote the integral constants and anisotropy constants in the inner and boundary parts of a grain, respectively. $\delta_B$ denotes the domain wall thickness. $M_s$ is the saturation magnetization, and $M_e$ in denominator of Equation (3) can be replaced by the saturation polarization $J_s$ in the International System of Units. $N_{\text{eff}}$ is the effective demagnetization factor.
Reference [12] considered that \( A' \) is equal to \( A \), and \( K_i' \) takes the fixed value less than \( K_i \). Based on our proposed anisotropy model, \( r_0 \) should be the thickness of anisotropic inhomogeneous district, and is denoted by \( r_0 \), and \( K_i' \) varies between 0 and \( K_i \). For convenience, \( K_i' \) in Equation (3) will be replaced by the average anisotropy \( <K_i'> \) in \( r_0 \) region. Thus, Equation (3) can be rewritten as,

\[
H_c = \frac{2K_i \pi r_0}{3\sqrt[3]{3M_s}} (\frac{A}{A} - \frac{(K_i')}{K_i}) - N_{eff} M_s
\]

where \( <K_i'> \) can be expressed as follows,

\[
<K_i'> = \left\{ \begin{array}{ll}
\frac{2}{\delta_0} \int_{r_0}^{d} (K_i - \Delta K (1 - \frac{2r - d}{2(d - r)})^3) dr + \int_{r_0}^{\infty} (K_i - \Delta K (1 - \frac{2r}{2})^3) dr,
\quad r_0 \leq \frac{lex}{2} \\
\frac{1}{r_0} \int_{r_0}^{\infty} (K_i - \Delta K (1 - \frac{2r - d}{2(d - r)})^3) dr + \int_{r_0}^{\infty} (K_i - \Delta K (1 - \frac{r}{2})^3) dr,
\quad \frac{lex}{2} \leq r_0
\end{array} \right.
\]

Taking the intrinsic magnetic parameters of \( \text{Nd}_2\text{Fe}_{14}\text{B} \):

\[
K_i = 4.3 \text{ MJ/m}^3, \quad A = 7.7 \times 10^{12} \text{ J/m}, \quad M_s = 1280 \text{ kA/m} \quad [13],
\quad J_f = 1.61 \text{ T} \quad [14], \quad \text{lex} = 4.2 \text{ nm} \quad [15], \quad \delta_0 = 4.2 \text{ nm}, \quad N_{eff} = 0.6 \quad [16],
\]

into Equations (4) and (5), we can calculate the coercivity of magnet for different values of \( r_0, d \) and \( K_i(0) \).

4. Results and Discussion

Figure 2 shows the variations of anisotropy \( K_i'(r) \) for given values of \( r_0d \) and \( K_i(0) \). For different values of \( r_0, d \) and \( K_i(0) \), \( K_i'(r) \) decreases with decreasing \( r \). This is due to that the closer to the grain surface, the smaller the anisotropy is [4]. It can be also seen that, for the fixed \( r_0 \) and \( K_i(0) \) shown by the star and circle lines, the variation velocities of \( K_i'(r) \) increases with increasing \( d \). This is attributed to the decreasing variation range from \( d/2 \) to \( r_0 \) with increasing \( d \) for the fixed value of \( (K_i - K_i(0)) \). But for the fixed \( r_0 \) and \( d \), shown by the upper triangle and lower triangle lines, the variation velocities of \( K_i'(r) \) decreases with increasing \( K_i(0) \), which is owing to that, the variation range \((K_i - K_i(0)) \) decreases with increasing \( K_i(0) \) for the fixed variation range from \( d/2 \) to \( r_0 \). While for the fixed \( d \) and \( K_i(0) \) shown by the circle and lower triangle lines, the variation speeds of \( K_i'(r) \) decreases with increasing \( r_0 \), attributing to the increasing variation range from \( d/2 \) to \( r_0 \) as increasing \( r_0 \) for the fixed value of \( (K_i - K_i(0)) \).

Figure 3 shows the dependence of average anisotropy, \( <K_i'> \), on \( d \) for different values of \( r_0 \) and \( K_i(0) \). For different \( r_0 \) and \( K_i(0) \), \( <K_i'> \) all decreases with increasing \( d \), which is attributed to the variation speeds of \( K_i'(r) \) increases with increasing \( d \) (as shown in Figure 2). So, \( <K_i'> \) computed by Equation (5) decreases. But for the fixed \( r_0 \) and \( d \) shown by the upper triangle and circle lines, \( <K_i'> \) increases with increasing \( K_i(0) \), which is owing to the variation velocities of \( K_i'(r) \) decrease with increasing \( K_i(0) \) (as shown in Figure 2). Thereby, \( <K_i'> \) calculated by Equation (5) increases. It can be also seen...
The increase of both the IP’s thickness \(d\) and GBSD’s thickness \(r_0\) or the decrease of the IP’s anisotropy constant \(K_1(0)\) all enhance the coercivity of magnet. Yet, if \(d\) and \(r_0\) are too larger and \(K_1(0)\) is too smaller, the magnetization and remanence would badly fall, then it is impossible to obtain high-energy product. In order to get high-energy product, it needs not only to enhance coercivity, but also to keep a sufficiently high remanence. Therefore, it is necessary to ensure that the IP’s thickness is around 1 nm, the GBSD’s thickness is around 6 nm, and \(K_1(0)\) varies between 0.1 \(K_1\) and 0.2 \(K_1\), by reasonably adjusting the alloy’s composition and technical process. So, this paper possesses a high preference value for experiment preparing high coercivity HDDR Nd-Fe-B magnet with considerable magnetization and remanence.

5. Conclusions

Effects of the IP’s thickness \(d\), its anisotropy constant \(K_1(0)\), and the GBSD’s thickness \(r_0\) on the coercivity in the HDDR Nd-Fe-B magnet are investigated. The results indicate that \(H_c\) increases with the increasing \(d\) and \(r_0\), but decreases with the increasing \(K_1(0)\). And while \(r_0=6\) nm, \(d=1\) nm and \(K_1(0)=0.15K_1\), the calculated coercivity is consistent well with experimental data.

7. Acknowledgements

The work is supported by the National Natural Science Foundation of China (50671055) and (50801043).

REFERENCES


