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Effect of MgO additive on coercivity, thermal stability and microstructure of Nd–Fe–B magnets

Aru Yan, Zhongmin Chen, Xiaoping Song, Xiaotian Wang

School of Materials Science and Engineering, Xi'an Jiaotong University, Xi'an 710049, People's Republic of China

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Abstract

The effect of MgO addition on the coercivity, thermal stability and microstructure of $Nd_{22}Fe_{71}B_7$ magnets has been investigated. Results show that both the coercivity and thermal stability of Nd–Fe–B magnets can be improved by the MgO additive. Microstructure studies reveal that a new intergranular Nd–O–Fe–Mg phase with a composition close to Nd_{70}–O_{22}–Fe_5–Mg_{1-3} appears in the magnets with MgO addition. The improvement of the properties of the magnets may be correlated with the appearance of this new phase. It was further found that the introduction of an appropriate amount of oxygen and Mg together is beneficial for the magnetic properties and thermal stability of Nd–Fe–B magnets.

Keywords: Nd–Fe–B; Permanent magnets; Additives; MgO; Sintering, Coercivity; Remanence

1. Introduction

Since the introduction of Nd–Fe–B magnets with superior permanent magnetic property at room temperature [1,2], many efforts have been made to improve both their properties and thermal stability [3–7]. The properties of magnets are dependent not only on the intrinsic magnetic properties of the main phase, but also on the characteristics, volume fraction and distribution of the intergranular phase [8,9]. So, it is possible to improve the properties of Nd–Fe–B magnets by an appropriate modification to the intergranular microstructure. Besenicar et al. reported that the coercivity and the temperature coefficients of the magnetic properties of sintered magnets can be improved with the addition of small amounts of ZrO_2 [10]. Kim and Camp reported that the addition of appropriate amounts of oxygen to an NdDyFeB magnet containing cobalt causes improvements in magnetic properties and thermal stability [11]. These results suggest that the addition of oxygen and some metallic elements together may be an effective approach to improve both the magnetic properties and thermal stability. In the present work, MgO was added to the intergranular region of Nd–Fe–B magnets and the effects on coercivity, thermal stability and microstructure were investigated.

2. Experimental

The magnets used in this study were prepared by the conventional powder metallurgy process, including induction melting the raw materials of 99% Nd, 99.99% Fe and 20%B–Fe, crushing and ball milling the ingots, pressing the powders under a magnetic field of 20 kOe, sintering the green compacts at 1000°C for 1 h. The as-sintered magnets were then heat treated at 625°C for 1 h. Fine MgO powder was added to the Nd–Fe–B powders prior to the ball milling. The alloy composition was $Nd_{22}Fe_{71}B_7$. The amount of MgO added was 2 wt.%.

Magnetic properties were measured using a DGY-2 hysteresisgraph. The specimens were examined with an MEF-3 optical microscope, an S-2700 scanning electron microscope (SEM) equipped with an energy-dispersive X-ray analysis (EDX) unit. The thermal stability was evaluated by the reversible loss of the open-circuit flux from room temperature up to 200°C.

3. Results and discussion

3.1. Magnetic property

Table 1 lists the remanence, coercivity and revers-

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Table 1
Remanence B_r, coercivity ${}_iH_c$ and reversible flux loss δ_{rc} from room temperature to 180°C for magnets with/without MgO and with Mg additions

Magnets	B_r (kGs)	${}_iH_c$ (kOe)	δ_{rc} (%, 180°C)
$Nd_{22}Fe_{71}B_7$	10.4	17.0	25.4
+2 wt.% MgO	9.5	22.1	20.5
+1.2 wt.% Mg	9.7	20.4	26.9

ible flux loss from room temperature to 180°C for magnets without (NFB) and with MgO addition (NFB-MO). It can be seen that the coercivity is considerably increased and the flux loss is decreased by the addition of MgO. Fig. 1 further shows the reversible flux loss as a function of the temperature for NFB and NFB-MO magnets. It reveals that up to 150°C the addition of MgO has almost no effect on the flux loss, but that the higher temperature, the greater the effect.

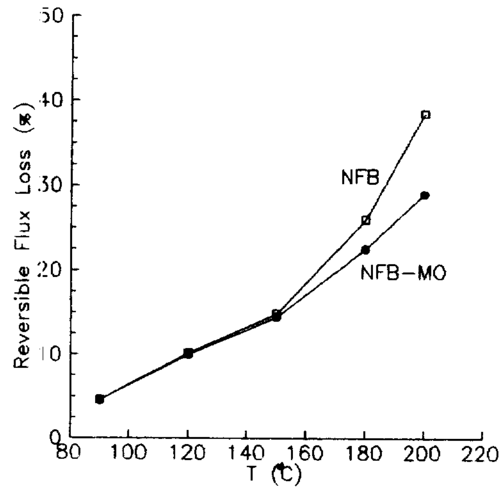

Fig. 1. Temperature dependence of reversible loss for magnets (a) without and (b) with MgO addition.

3.2. Microstructure

Microstructural studies show that the $Nd_2Fe_{14}B$ matrix of the magnets remains unchanged but that the intergranular microstructure varies markedly with MgO addition. The SEM micrographs for NFB and NFB-MO magnets are shown in Fig. 2. Table 2 lists the composition of the intergranular phase, which was determined by an EDX unit. It can be seen that there is an Fe–Nd–O net-like phase with a composition close to Fe_{71}–Nd_{26}–O_3 in the intergranular regions of NFB magnets. However, in the NFB-MO magnets, the volume fraction of the net-like phase greatly decreases and a new particle-like intergranular phase appears. This new phase has a composition close to Nd_{70}–O_{22}–Fe_5–Mg_{1-3}. By SEM observation and EDX analysis no MgO particles were found in the intergranular regions of NFB-MO magnets. Therefore, we believe that this new Nd–O–Fe–Mg phase is formed via a reaction between MgO and the intergranular Fe–Nd–O phase during the sintering.

3.3. Effect of the Nd–O–Fe–Mg phase on the coercivity and thermal stability

The above microstructural studies suggest that the improvement of the coercivity and thermal stability should be correlated with this new phase. It has a lower Fe content and thus a lower magnetic permeability. This may reduce the magnetic coupling of

Table 2
Composition of the intergranular phase in magnets with/without MgO and with Mg additions

Magnets	Nd (at.%)	Fe (at.%)	O (at.%)	Mg (at.%)
$Nd_{22}Fe_{71}B_7$	26.07	71.30	2.63	–
+2 wt.% MgO	70.26	5.04	22.87	1.82
+1.2 wt.% Mg	36.51	60.26	2.46	0.77

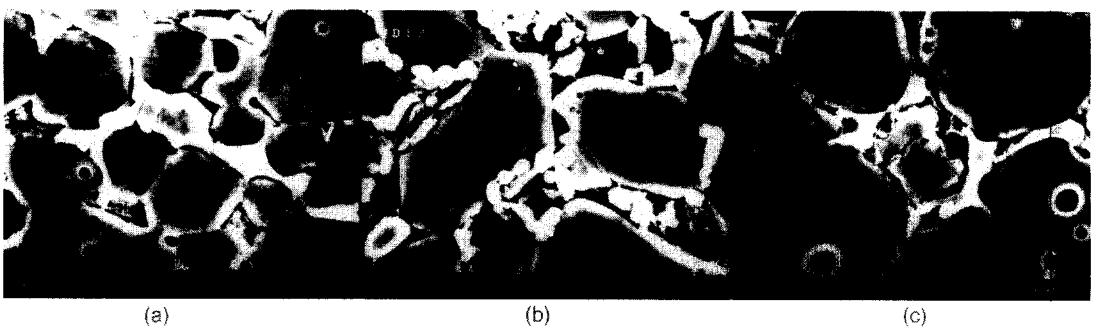

Fig. 2. SEM micrographs for magnets (a) without, (b) with MgO and (c) with Mg addition.

the $Nd_2Fe_{14}B$ grains. In view of the particle-like morphology of the Nd–O–Fe–Mg phase in the $Nd_2Fe_{14}B$ grain boundaries, we believe that the propagation of the domain walls from grain to grain is impeded effectively by this new phase, which enhances the coercivity. Owing to the lower Fe content, the magnetic characteristic of this new phase is not sensitive to temperature. It is supposed that the improvement of the thermal stability at high temperature can be attributed to this. However, the mechanism of this effect is not clear and further investigations are needed.

3.4. The role of oxygen

In order to investigate the role of oxygen, which is a constituent element of MgO, pure elemental Mg was added to the intergranular regions of Nd–Fe–B magnets. The amount of the addition is 1.2 wt.%, which is equal in mass to the amount of the elemental Mg that may have formed from the 2 wt.% MgO addition. The results are shown in Tables 1 and 2 and Fig. 2. It can be seen that the coercivity is also enhanced, but slightly less than that of the NFB-MO magnets. The flux loss is slightly higher than that of NFB magnets. SEM observation and EDX analysis show that an intergranular net-like Fe–Nd–O–Mg phase with a composition close to Fe_{50-66}–Nd_{30-40}–O_{2-5}–Mg_{2-3} appears in the magnets with Mg addition. It is obvious that this phase is similar to the Fe–Nd–O phase in the NFB magnets and different from the Nd–O–Fe–Mg phase in the NFB-MO magnets.

The above studies reveal that the effects of the element Mg are inferior to those of the oxide MgO. It has been reported that oxygen is detrimental to the coercivity and thermal stability of magnets [12,13]. These results reveal that the single addition of Mg or O cannot effectively improve the properties of Nd–Fe–B magnets, but the introduction of Mg and O together is beneficial.

4. Conclusions

(1) Intergranular addition of MgO can modify the coercivity, thermal stability and intergranular microstructure of sintered Nd–Fe–B magnets.

(2) Owing to the reaction between MgO and the intergranular Fe–Nd–O phase, a new intergranular Nd–O–Fe–Mg phase is formed in the magnets with MgO addition. The improvement of the coercivity and the thermal stability can be correlated with the occurrence of this new phase.

(3) Oxygen can be helpful to improve the properties of Nd–Fe–B magnets when added to the intergranular regions together with Mg.

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References

[1] M. Sagawa, S. Fujimura, N. Togawa et al., *J. Appl. Phys.*, *55* (1984) 2083.
[2] J. Croat, J. Herbst, R.W. Lee et al., *J. Appl. Phys.*, *55* (1984) 2078.
[3] M. Sagawa, H. Kogure, M. Endoh et al., *IEEE Trans. Magn.*, *20* (1987) 1594.
[4] M. Tokunaga, H. Kogure, M. Endoh et al., *IEEE Trans. Magn.*, *23* (5) (1987) 2287.
[5] A.S. Kim, *J. Appl. Phys.*, *63* (1988) 3519.
[6] A.S. Kim, *J. Appl. Phys.*, *63* (1988) 3975.
[7] X. Shen, Y. Wang, Z. Diao et al., *J. Appl. Phys.*, *61* (1987) 3433.
[8] J. Fidler, *IEEE Trans. Magn. 23* (1987) 2106.
[9] J.M. Elbicki, W.E. Wallace and V. Korablev, *IEEE Trans. Magn. 25*, (1989) 3567.
[10] S. Besenicar, B. Saje, G. Drazic et al., *J. Magn. Magn. Mater.*, *104–107* (1992) 1175.
[11] A.S. Kim and F.E. Camp, personal communications.
[12] A.S. Kim, *J. Appl. Phys.*, *64* (1988) 5571.
[13] T. Minowa, M. Shimao and M. Honshima, *J. Magn. Magn. Mater.*, *97* (1991) 107.