The Effect of Coupling Agent on the Microwave Properties of the Melt-Spun Iron/Earth Nanocomposites

Xie Guozhi,[1,2] Wang Ping,[1] Yuan Liukui,[1] Zhang Baoshan,[2] Lin Pinghua,[1] Lu Huaixian[2]

[1]*College of Materials Science and Engineering, Hohai University, Nanjing 210098, People's Republic of China*
[2]*Department of Physics, Nanjing University, Nanjing 210093, People's Republic of China*

Received 19 January 2009; accepted 25 May 2009
DOI 10.1002/app.30831
Published online 2 July 2009 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The effects of silane coupling agent doping on the microstructure and microwave properties of melt-spun Nd-Fe-Co alloys were investigated. All as-milled powders, with or without silane coupling agent treatment, were characterized as flake-like shape. However, it is found that with the addition of 0.25 mL coupling agent, the as-milled Nd-Fe-Co powders can deagglomerate and become more uniform dispersion with smaller thickness. In contrast, the excessive addition of coupling agent led to somewhat higher thickness of the flakes and much more amount of coupling agent on the surface of powder. The electromagnetic properties show that the Reflection loss will arrive −27.1 dB at 4.2 GHz, for the addition of 0.25 mL coupling agent powders. The probably reasons for this are discussed on the basis of the morphology of the powders, uniform dispersion and the physical coating between the polymer and coupling agent. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 114: 2344–2347, 2009

Key words: microstructure; alloys; matrix

INTRODUCTION

Recently, the electromagneticwave (EM) absorbing materials have been attracting much attention because the electromagnetic interference problems become more common along with the rapid development and spread of electronic devices using the electromagnetic wave in GHz range.[1] Therefore, the polymer composites containing a large amount of the magnetic-metal particles have been used as EM absorber.[2] In practice, the wettability and compatibility can be greatly reduced due to the chemical heterogeneity and/or the physical roughness of the metal surfaces. This is turn will result in imperfect couplings, which form voids in the bonded magnet. To overcome these obstacles, one approach is to introduce a "bridge," between the metal surface and the binder.[3] Li et al.[4] found that the coupling treatment can improve the interfaces between the powders and the binders, decrease in volume fraction of the binder increases magnetic properties of the bonded NdFeB magnet. Duan et al.[5] found that the addition of 10 wt % Titanate coupling agent to the filled carbon black (CB) improved the microwave absorption performance of the CB/acrylonitrile-butadiene-styrene composites. However, the effect of coupling agent on the microstructure and EM properties of melt-spun iron/earth is seldom studied. In this article, the surface modification, dispersion, and microwave properties of the melt-spun Nd-Fe-Co powders with or without doping coupling agent were studied. The purpose of this article is to study the effect of coating treatment on the microstructure and EM properties of melt-spun iron/earth powders.

EXPERIMENTAL

The $Nd_4(Fe_{0.7}Co_{0.3})_{96}$ alloys were prepared using metals in purity of Nd 99.8 wt %, Co 99.5 wt %, and Fe 99.5 wt %, respectively. These ingots were initially alloyed by arc melting for three times to obtain homogeneous composition and melt-spun in a molybdenum wheel in argon atmosphere. The wheel speeds of 30 m/s were used. The ribbons were first crushed by a crusher for 20 min. Then the primary powders with acetone as medium were loaded with multiple diameter steel ball in a steel container with ball-to-powder weight ratio of 10 : 1. The mechanical milling with 500 rpm for 2 h (as-milling powder). At last the silane coupling agent (KH-560) was added into as-milling powders (5 g) with 0, 0.25, 0.5, and 1 mL, respectively. Mixed with appropriate alcohol, these powders mixture

Correspondence to: X. Guozhi (guozhixie@yahoo.com.cn).
Contract grant sponsor: Postdoctoral Support Program in Scientific Research of Jiangsu Province; contract grant number: 0204003425.
Contract grant sponsor: Cyanine Project of Jiangsu Province (2006).

© 2009 Wiley Periodicals, Inc.

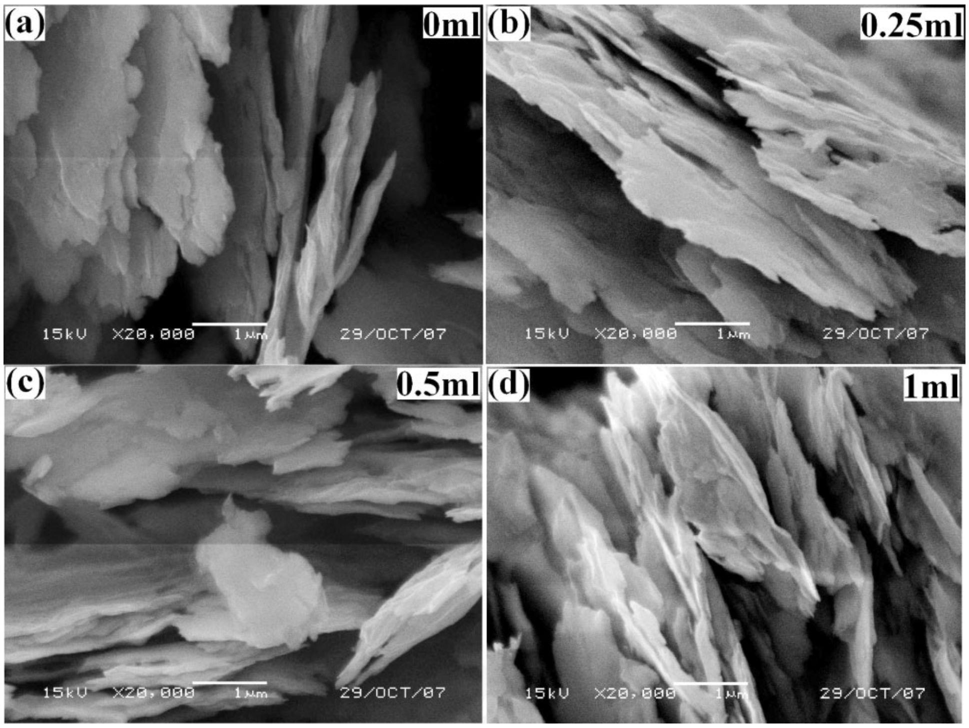

Figure 1 SEM micrographs of the powders with different content of coupling agent after milling(a) 0 mL; (b) 0.25 mL; (c) 0.5 mL; and (d) 1 mL.

were milled with vibrating cup for 1 h. The microstructure was detected by scanning electron microscope (SEM). Composite materials were prepared by dispersing the powders in paraffin wax, and the weight fraction of powder is 70%. The powder wax composites were die-pressed to form cylindrical toroidal specimens with 7.0 mm outer diameter, 3.04 mm inner diameter, and 3–6 mm thickness. The measurements of complex permeability μ and permittivity ε for the specimens were carried out using an Agilent E8363B vector network analyzer in the 2–18 GHz ranges.

RESULTS AND DISCUSSION

Figure 1 shows the SEM micrographs of the powders with different doping content of coupling agent after milling. It can be seen that all the powders were deformed into flakes with an increase of the plan size (length) and a decrease of thickness. It is well known that ferromagnetic metallic thin flakes have high magnetic permeability in gigahertz frequencies above snoek's limit due to their large saturation magnetization and the effects of particle shape.[6,7] From Figure 1(a), without coupling agent doped, the milled powders have less uniform dispersion and more agglomeration with somewhat larger thickness. However, it can be observed that the milled powders with doped 0.25 mL coupling agent, as shown in Figure 1(b), have well uniform dispersion and deagglomeration, with thin thickness, as shown in Figure 1(b). It will be favorable to get excellent microwave properties with higher aspect ratio.[8,9] With the addition of 0.5 and 1 mL coupling agent, as shown in Figure 1(c) and Figure 1(d), the thickness of the flake-shape powders seem somewhat augment and much more coupling agents were absorbed on the surface of the flakes. Thus, the improved doped content of coupling agent may be disadvantage to the microwave properties because the amount of added filler decreased.[10]

Figure 2 shows the frequency dependence of the complex permeability for milled samples with different coupling agent content. Both samples have the largest μ' at 2 GHz and decrease smoothly with frequency. It is noted that the powders with 0.25 mL coupling agent addition has highest μ', about 3.3, while the μ'' value of it has dramatically increased greatly in comparison to that with other samples. The μ'' of powders arrives 2.5 at 12.7 GHz with the addition of 0.25 mL coupling agent, whereas that of other powders is lower than 1.4. This may be due to the excessive amount of doping coupling agent decrease the filling density of magnetic phases.

Figure 3 shows the frequency dependence of the complex permittivity. In general, it can be seen that

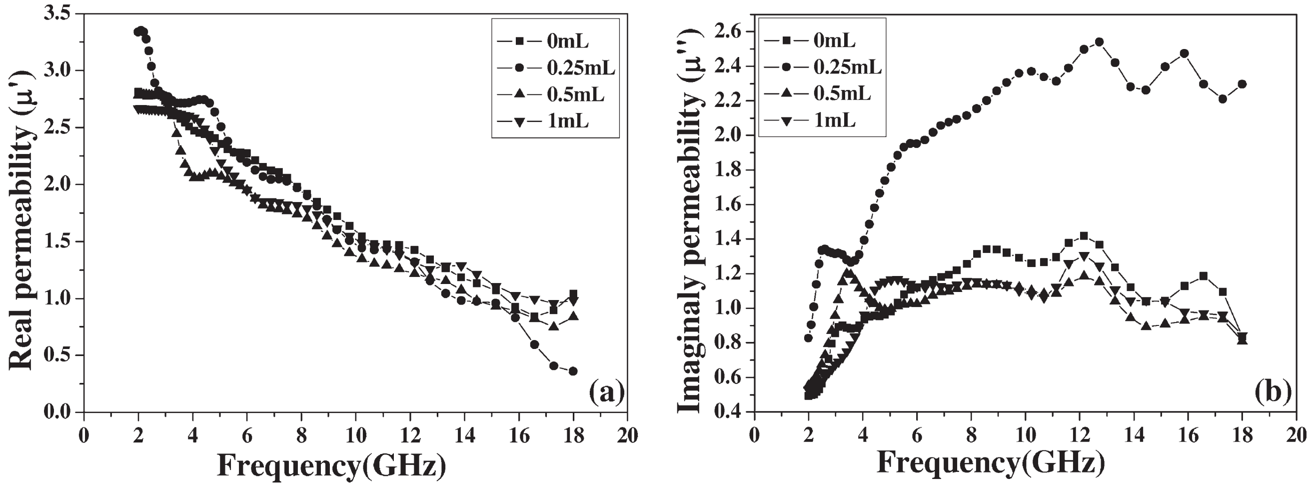

Figure 2 Complex permeability of milled samples with different coupling agent content.

for both samples with coupling agent doped, the ε' values remain smooth decreased with the frequency, whereas the ε'' values remain increased. It is noted that the powders without of coupling agent addition have the highest ε' and ε'' values. However, the ε' and ε'' values decreased with the content of doping coupling agent increased. The reason should be attributed to the metal powders coated by the coupling agent were isolated from each other.[5] However, the effect of that becomes weaker and weaker with higher content of doping coupling agent for the isolated effect of powders arrived maximum.

According to the transmission line theory, the reflection loss (RL) of normal incident electromagnetic wave at the absorber surface can be calculated from the relative permeability and permittivity at a given frequency and absorber thickness using the following equations[6]:

$$Z_0 = \sqrt{\frac{\mu_0}{\varepsilon_0}},$$

$$Z_{\mathrm{in}} = \sqrt{\frac{\mu}{\varepsilon}}\tanh\left(j\frac{2\pi fd}{c}\right)\sqrt{\mu\cdot\varepsilon}, \text{ and}$$

$$RL = 20\lg\left|\frac{Z_{\mathrm{in}} - Z_0}{Z_{\mathrm{in}} + Z_0}\right|.$$

Here, Z_0 is the impedance of air, μ_0, ε_0 are the permeability and permittivity of air, respectively, f is the frequency of the electromagnetic wave, d is the thickness of the absorber, c is the velocity of light, Z_{in} is the input impedance of the absorber, and μ, ε are the complex permeability and complex permittivity of absorber, respectively.

Figure 4(a) shows the frequency dependence of the calculated RL for different coupling agent content after milling with the thickness of 1.5 mm. It

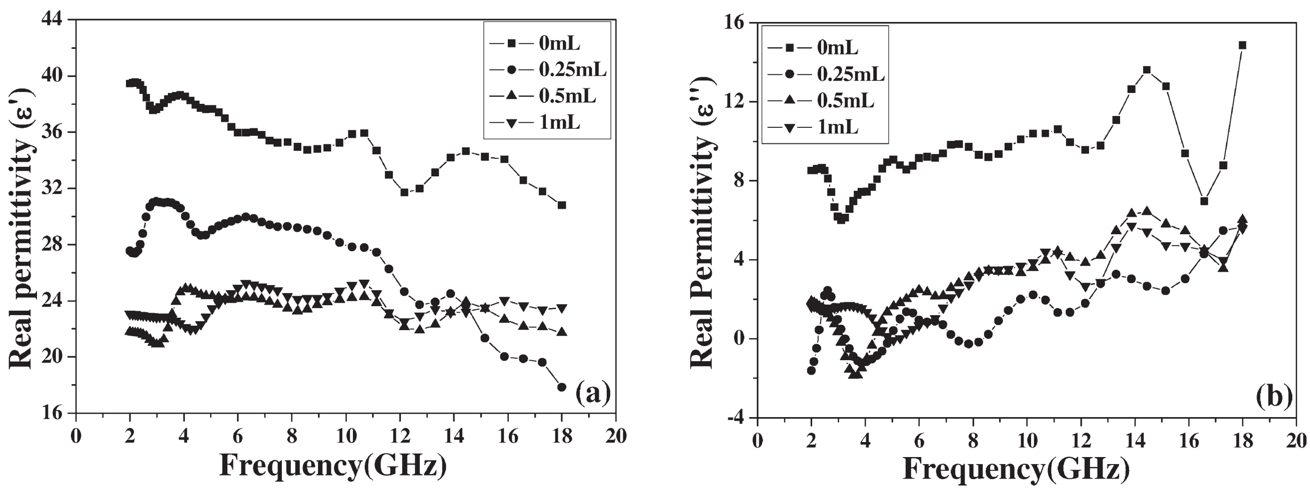

Figure 3 Complex permittivity of milled samples with different coupling agent content.

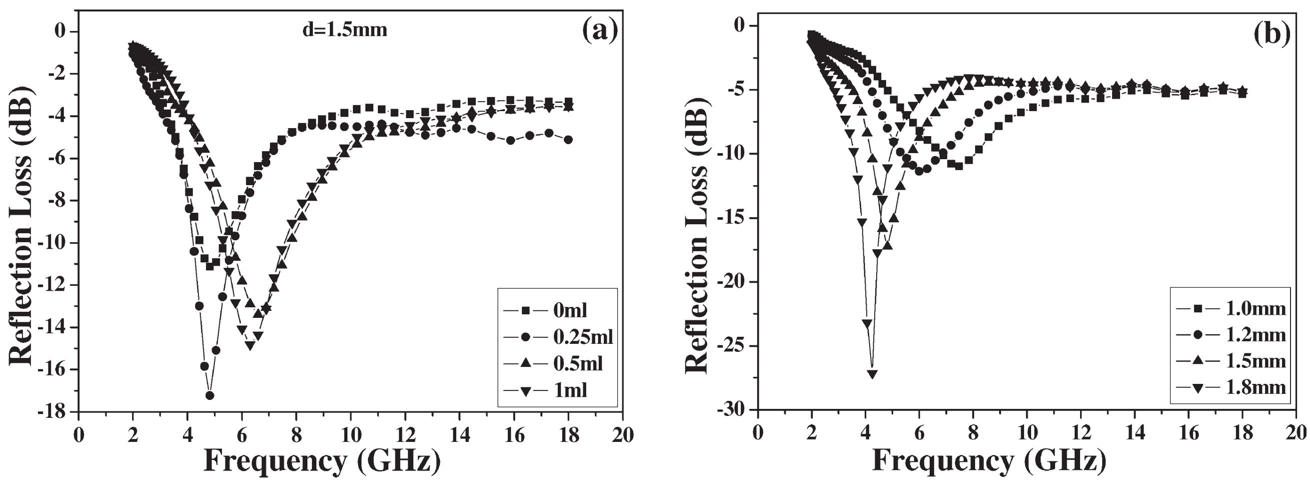

Figure 4 Reflection loss (RL) of the milled powders (a) powders with different coupling agent content; (b) powders with addition of 0.25 mL with different thickness.

shows that with the content of doping coupling agent increased, the intensity of RL increased, whereas the bandwidth decreased. However, the position of RL peaks is shifted to lower frequency. The minimal loss is −17.2dB at 4.8 GHz for the powders with addition content of 0.25 mL coupling agent. Figure 4(b) shows the powders with addition content of 0.25 mL coupling agent with different thickness, 1, 1.2, 1.5, and 1.8 mm, respectively. The minimal loss is −27.1 dB at 4.2 GHz with the thick 1.8 mm. Excellent microwave absorbing properties of powders should be due to the dispersed magnetic-metal particles in the matrix were coated by the coupling agent and were isolated from each other. Furthermore, some magnetic-metal particles were embraced by coupling agent and formed isolated conducting aggregates. The contactless (isolated) distribution of the particles in the paraffin wax matrix led to the improvement of the absorbing and reflection activities.[5] However, the low permittivity was obtained with higher doping content of coupling agent. This will cause the lower effect volume of magnetic-metal. So the low permeability was obtained and led to low RL for higher addition content of coupling agent.

CONCLUSIONS

The addition of silane coupling agent to melt-spun Nd-Fe-Co composites had a notable influence on the microstructure and microwave properties. With the addition of 0.25 mL coupling agent, the powders can deagglomerate and more uniform dispersion with smaller thickness, whereas the larger addition content of coupling agent led to somewhat higher thickness. The reason for this are that the coupling agent improved adhesion between the metal powders and paraffin wax, which can led to uniform dispersion of the metal powders.

References

1. Li, D.; Horikawa, T.; Liu, J. R.; Itoh, M.; Machida, K.-I. J Alloys Compd 2006, 408, 1429.
2. Choi, H.; Moon, K.; Jeon, I.; Kim, W.; Moon, T. J Appl Polym Sci 1997, 66, 477.
3. Chen, Q.; Asuncion, J.; Landi, J.; Ma, B. M. J Appl Phys 1999, 85, 5684.
4. Li, J.; Liu, Y.; Gao, S. J.; Li, M.; Wang, Y. Q.; Tu, M. J. J Magn Magn Mater 2006, 299, 195.
5. Duan, Y.; Liu, S.; Wang, G.; Guan, H.; Wen, B. J Appl Polym Sci 2006, 102, 1839.
6. Zhang, B.; Lu, G.; Feng, Y.; Xiong, J.; Lu, H. J Magn Magn Mater 2006, 299, 205.
7. Wang, X.; Gong, R.; Li, P.; Liu, L.; Cheng, W. Mater Sci Eng A 2007, 466, 178.
8. Qiao, L.; Wen, F.; Wei, J.; Wang, J.; Li, F. J Appl Phys 2008, 103, 063903.
9. Deng, L. J.; Zhou, P. H.; Xie, J. L.; Zhang, L. J Appl Phys 2007, 101, 103916.
10. Choi, H. D.; Kim, W. S.; Joon, I. S.; Yoon, H. G.; Moon, T. H. Polymer (Korea) 1997, 21, 112.