

Contents lists available at ScienceDirect

Journal of Alloys and Compounds



journal homepage: www.elsevier.com/locate/jallcom

Review

Epoxy resin effect on anisotropic Nd-Fe-B rubber-bonded magnets performance

Fuqiang Zhai^a, Aizhi Sun^{a,*}, Duo Yuan^a, Jin Wang^a, Shen Wu^a, Alex A. Volinsky^b, Zhixin Wang^b

^a School of Material Science and Engineering. University of Science and Technology Beijing, Beijing 100083. China ^b Department of Mechanical Engineering, University of South Florida, Tampa FL 33620, USA

ARTICLE INFO

Article history: Received 6 July 2010 Received in revised form 30 September 2010 Accepted 30 September 2010 Available online 12 October 2010

Keywords: Epoxy resin Lubricant Magnetic alignment Anisotropy Nd-Fe-B flexible bonded magnets

ABSTRACT

A novel anisotropic Nd–Fe–B flexible bonded magnet with epoxy resin lubricant was prepared by the two-step method to enhance its performance. Temperature characteristics of epoxy resin and its effect on magnetic properties and preparation of anisotropic Nd-Fe-B flexible bonded magnets were investigated and optimized. DOA of aligned flexible bonded magnets with epoxy resin lubricant increases significantly due to epoxy resin lower viscosity and subsequent better powder particles lubrication at a certain aligning temperature. Meanwhile, H_{irr} decreases sharply due to improved oxidation resistance of epoxy resin fully encapsulating magnetic powder during magnetic alignment process. Utilizing 1 wt% optimized encapsulating epoxy resin and heating unaligned flexible bonded magnets to 80 °C for 30 min during magnetic alignment resulted in the largest $\Delta(BH)_{max}$ and Δ DOA. $\Delta(BH)_{max}$ increased to over 126% along with Δ DOA increase to over 75%, much higher compared with unaligned flexible bonded magnets prepared exclusively by calendering.

© 2010 Elsevier B.V. All rights reserved.

۰.	n	te	n	Ľ

S

1.	Introduction	687
2.	Experimental details	688
3	Results and discussion	688
3. 4.	 3.1. Epoxy resin viscosity temperature dependence	688 688 688 689 689 689 690 690

1. Introduction

Rubber-bonded Nd–Fe–B is a new type of flexible magnets. Isotropic Nd-Fe-B ring-shaped rubber-bonded magnets have been used in small motors and actuators for personal computers and peripherals. However, computers miniaturization inevitably stresses the importance of flexible bonded magnets with superior magnetic properties contributing to the overall computer size reduction [1–11].

During magnetic alignment powder particles rotate along the easy magnetization direction due to externally applied magnetic field. Larger number of aligned particles results in better magnetic properties. Our group has developed a novel two-step method which includes calendering and magnetic alignment steps with heating to prepare anisotropic Nd–Fe–B flexible bonded magnets. Heating unaligned flexible bonded magnets to 100 °C for 30 min during magnetic alignment resulted in the largest $\Delta(BH)_{max}$, which increased by over 60% along with Δ DOA increase by over 39% compared with unaligned flexible bonded magnets prepared exclusively by calendering. However, binder viscosity increases due to rubber cross-linking when heated, which inevitably hinders further improvements in aligned flexible bonded magnets magnetic properties that originate from larger powder particles rotational resistance, *T*, during magnetic alignment: $T \propto \tau = \eta \gamma$, where τ is the shear stress between the powder particles and the binder, η is the binder viscosity and γ is the rotational rate of the powder particles. High binder viscosity prevents powder particles rotation during magnetic alignment.

A novel anisotropic Nd-Fe-B flexible magnet bonded with epoxy resin lubricant was prepared by the two-step method. One

^{*} Corresponding author. Tel.: +86 10 82376835; fax: +86 10 62333375. E-mail address: sunaizhi@126.com (A.Z. Sun).

^{0925-8388/\$ -} see front matter © 2010 Elsevier B.V. All rights reserved. doi:10.1016/j.jallcom.2010.09.210



Fig. 1. Process for preparing anisotropic Nd-Fe-B based flexible boned magnets with epoxy resin lubricant.

of the advantages of adding epoxy resin lubricant is that rotational resistance of magnetic powders decreases rapidly. Subsequently powder particles with easy magnetized directions rotate smoothly along the aligning direction as a result of the epoxy resin lower binder viscosity when heated to a certain temperature. Another advantage of adding epoxy resin lubricant is that flexible bonded magnets have higher strength compared with flexible magnets bonded with conventional binders at room temperature [12–25].

2. Experimental details

Two-step method flow chart for preparing anisotropic Nd-Fe-B flexible bonded magnets with epoxy resin lubricant is shown in Fig. 1. First bisphenol-A epoxy resin (0 wt%, 0.5 wt%, 1 wt%, 1.5 wt%) including diethylenetriamine (DTA) curing agent was dissolved in solution with the appropriate amounts of acetone. Then 100 mesh anisotropic HDDR Nd-Fe-B powders ($B_r = 1.22$ T, $H_{ci} = 930$ kA/m and $(BH)_{max} = 248 \text{ kJ/m}^3$) were placed into these four solutions. When acetone completely evaporated, four kinds of epoxy resins that fully encapsulated Nd-Fe-B powders were obtained. The second step is the calendering process, which consists of the following procedures. Initially four magnetic powders with KH550 silane coupling agent were dry-blended separately with 4.8 wt% chlorinated polyethylene (CPE) and plasticizer (Di-n-octyl phthalate). Then they were sheet-rolled individually to 1.5–2.0 mm with a twin roll-mill at 70–90 $^\circ$ C in N₂ atmosphere. The next magnetic alignment process step with heating consisted of the following procedures. First, the four kinds of flexible sheet samples were heated to 50-160 °C for 10-40 min. Then one part of samples of each kind was immediately placed into a pulsed magnetic field (3600 kA/m) for alignment. Other samples were left without any magnetic treatment. Finally all samples were cooled to room temperature. Epoxy resin viscosity was measured with an RS600 rotational rheometer (Rheology Solutions, Australia). Magnetic properties of all samples were measured after magnetization with a vibrating sample magnetometer (VSM) and a B-H curve tracer, respectively, in a pulsed magnetic field of 3600 kA/m. The rate of degree of alignment change, Δ DOA, was calculated as Δ DOA = (B_T – B₀)/B₀, where B_T and B₀ are remanence values of the aligned and unaligned samples parallel to the applied magnetic field, respectively. The magnetic energy product rate change, $\Delta(BH)_{max}$, was calculated as $\Delta(BH)_{max} = [(BH)_{max(T)} - (BH)_{max(T_0)}]/(BH)_{max(T_0)}$, where $(BH)_{max(T)}$ and $(BH)_{max(T_0)}$ are energy product values of the aligned and unaligned samples, respectively. The magnetic irreversibility field, Hirr, indicates the irreversible loss of energy product of bonded magnets during heating. H_{irr} was calculated as H_{irr} =



Fig. 2. Epoxy resin viscosity change at various temperatures.

 $[(BH)_{max(T_0)} - (BH)_{max(T^*)}]/(BH)_{max(T_0)}$ [26], where $(BH)_{max(T^*)}$ is the energy product of the unaligned samples heated at the same conditions as the aligned samples.

3. Results and discussion

3.1. Epoxy resin viscosity temperature dependence

Fig. 2 shows epoxy resin viscosity variations with time at different test temperatures. When heated to a certain temperature, epoxy resin viscosity decreases significantly and reaches the minimum value within 1–2 min. However, it increases gradually with time, being a function of the curing agent. When unaligned flexible bonded magnets with epoxy resin lubricant are heated to a certain magnetic aligning temperature, rotational resistance of powder particles reduces due to the lower binder viscosity, therefore, good alignment results can be achieved. Consequently, epoxy resin can be used as a lubricant and fully encapsulate magnetic powder to prepare anisotropic Nd–Fe–B flexible bonded magnets by the twostep method.

3.2. Epoxy resin content effect on magnetic properties of anisotropic Nd–Fe–B flexible bonded magnets

Fig. 3 shows $\Delta(BH)_{max}$ and Δ DOA temperature dependence for aligned samples encapsulated by different epoxy resins. $\Delta(BH)_{max}$ and Δ DOA of aligned samples with epoxy are greater than those without epoxy at each aligning temperature, indicating that adding appropriate amount of epoxy resin improves magnetic properties of the aligned samples. As a result, the largest $\Delta(BH)_{max}$ of the aligned samples increases to 126.7% from 60.5% by adding 1 wt% epoxy resin, and the largest Δ DOA increases from 49.5% to 75%.

As seen in Fig. 3, when epoxy resin content is 1 wt%, both $\Delta(BH)_{max}$ and Δ DOA reach the maximum at each aligning temperature. This phenomenon can be explained as follows. When epoxy resin content is higher than 1 wt%, magnetic properties of the aligned samples begin to decrease due to the non-magnetic nature and magnetic dilution of epoxy resin. When epoxy resin content is lower than 1 wt%, magnetic properties also decline, since powder particles cannot be encapsulated sufficiently by epoxy resin, resulting in reduced lubrication and subsequent difficulty in particles relative rotation. As a result, epoxy resin is used as a lubricant to improve magnetic properties of anisotropic Nd–Fe–B flexible boned magnets, and the optimal epoxy resin content is 1 wt%.



Fig. 3. Magnetic properties temperature dependence for samples encapsulated by different epoxy resins.

3.3. Epoxy resin lubricant effect on the magnetic alignment process with heating

When epoxy resin content is 1 wt% magnetic properties of the aligned samples are the best at each aligning temperatures. Thus this optimal 1 wt% content was selected to study the effects of adding epoxy resin lubricant on the magnetic alignment process with heating.

Fig. 4 shows $\Delta(BH)_{max}$, Δ DOA and H_{irr} variations with temperature for aligned samples. Both $\Delta(BH)_{max}$ and Δ DOA exhibit a similar change with temperature. In the 60–80 °C range they both increase rapidly. Moreover, the largest $\Delta(BH)_{max}$ of 126.7% and the largest Δ DOA of 75% were obtained at 80 °C during magnetic alignment process with heating. On the contrary, magnetic properties degrade gradually above 80 °C and sharply above 100 °C.

It is useful to shorten the holding time to reduce $H_{\rm irr}$. Meanwhile, the largest Δ DOA of 75% was obtained at 80 °C. Therefore, changes in magnetic properties of aligned samples were investigated as a function of holding time at 80 °C shown in Fig. 5. The largest Δ DOA of 75% was obtained at 80 °C held for 30 min. Consequently, the largest Δ (*BH*)_{max} of 126.7% for aligned samples was obtained after 30 min holding time.



Fig. 4. Changes in magnetic properties as a function of temperature.



Fig. 5. Changes in magnetic properties as a function of holding time.

The effect of temperature and holding time on DOA of the aligned samples was analyzed from the standpoint of the epoxy resin viscosity change with temperature (Fig. 2). As seen in Fig. 2, viscosity of heated epoxy resin decreases significantly and reaches the minimum within 2 min, which is not useful for preparing flexible bonded magnets by the two-step method due to short holding time. When heated and held at 120 °C, viscosity reaches the minimum for a short time among these heating temperatures, but it increases almost linearly above 6 min due to curing. Therefore, in the 60–130 °C range, when holding time is above 6 min, viscosity has the minimum at 80 °C, which explains why the largest Δ DOA appears at 80 °C.

As seen in Fig. 4, when temperature is elevated from $60 \,^{\circ}$ C to $80 \,^{\circ}$ C, Δ DOA increases continuously, while H_{irr} is very low and unchanging. This is clearly illustrated by improved magnetic properties of the aligned samples, and $\Delta(BH)_{max}$ increases significantly with increasing temperature from $60 \,^{\circ}$ C to $80 \,^{\circ}$ C. The best magnetic properties of the aligned sample were obtained, which exhibited the largest $\Delta(BH)_{max}$ increase to 126.7% by means of magnetic alignment at $80 \,^{\circ}$ C. When temperature reaches $80 \,^{\circ}$ C or above, Δ DOA declines gradually with temperature. Meantime, H_{irr} increases slightly with temperature. This shows that magnetic properties of the aligned samples begin to deteriorate gradually, and $\Delta(BH)_{max}$ decreases. Δ DOA significantly decreases, and H_{irr} rises rapidly as heating temperature is elevated above $100 \,^{\circ}$ C. Therefore, magnetic properties of the aligned samples decline sharply.

In conclusion, when selecting the best encapsulating epoxy resin content of 1 wt%, the optimal preparation condition is heating unaligned flexible bonded magnets to $80 \,^{\circ}$ C for $30 \,$ min for magnetic alignment.

3.4. Epoxy resin lubricant effect on the magnetic irreversibility field of anisotropic Nd–Fe–B flexible boned magnets

Fig. 6 shows H_{irr} variations with temperature for anisotropic Nd–Fe–B flexible boned magnets with and without epoxy resin. When epoxy resin content is 1 wt%, H_{irr} of the aligned samples is less than that of magnets without epoxy resin encapsulation at each aligning temperature, with a larger difference above 100 °C. When epoxy resin fully encapsulates powder particles, it prevents their oxidation and helps increase energy product of anisotropic Nd–Fe–B flexible boned magnets during magnetic alignment process with heating.



Fig. 6. Temperature dependence of the magnetic irreversibility field for anisotropic Nd–Fe–B flexible boned magnets with different epoxy resin contents.

4. Conclusions

Adding appropriate amount of epoxy resin lubricant helps prepare anisotropic Nd–Fe–B flexible boned magnets with superior magnetic properties. Adding epoxy resin plays lubrication role to weaken powder particles rotational resistance and subsequently increase aligned samples Δ DOA when heated to a certain aligning temperature. Meanwhile, adding epoxy resin plays an important role in preventing powder oxidation and increasing $\Delta(BH)_{max}$ of the aligned samples during magnetic alignment process with heating. When epoxy resin content is 1 wt%, aligned samples $\Delta(BH)_{max}$ and Δ DOA reach the maximum at each aligning temperature. Thus 1 wt% epoxy is the optimal encapsulating content. When selecting the best encapsulating content of 1 wt%, the optimal preparation condition is heating unaligned flexible bonded magnets to 80 °C for 30 min for magnetic alignment. $\Delta(BH)_{max}$ and Δ DOA of anisotropic Nd–Fe–B flexible bonded magnet increase to 126.7% and 75% compared with unaligned flexible bonded magnets prepared exclusively by calendering.

References

- S. Hirosawa, H. Kanekiyo, T. Miyoshi, Y. Shigemoto, K. Murakami, Y. Senzaki, T. Nishiuchi, J. Alloys Compd. 408 (2006) 260.
- [2] D. Plusa, B. Slusarek, M. Dospial, U. Kotlarczyk, T. Mydlarz, J. Alloys Compd. 423 (2006) 81.
- [3] F. Yamashita, M. Nakano, H. Fukunaga, J. Magn. Magn. Mater. 303 (2006) 363.
 [4] F. Yamashita, M. Ogushi, M. Nakano, H. Fukunaga, J. Magn. Magn. Mater. 310
- (2007) 2578. [5] L.X. Hu, A.J. Williams, I.R. Harris, J. Alloys Compd. 460 (2008) 232.
- [6] P. Saravanan, A.N. Sharma, V. Chandrasekaran, J. Magn. Magn. Mater. 321 (2009) 3138.
- [7] X.K. Lv, S.W. Or, W. Liu, X.H. Liu, Z.D. Zhang, J. Alloys Compd. 476 (2009) 271.
- [8] S. Li, S.X. Bai, H. Zhang, K. Chen, J.Y. Xiao, J. Alloys Compd. 470 (2009) 141.
- [9] F. Hirotoshi, M. Hiroki, Y. Takeshi, N. Masaki, Y. Fumitoshi, J. Appl. Phys. 107 (2010) 736.
- [10] T. Saito, J. Alloys Compd. 505 (2010) 23.
- [11] H.W. Chang, M.F. Shih, C.C. Hsieh, W.C. Chang, C.Y. Shen, J. Alloys Compd. 489 (2010) 499.
- [12] F. Yamashita, Y. Yamagata, H. Fukunaga, IEEE Trans. Magn. 36 (2000) 3366.
- [13] O. Mauerer, Polym. Degrad. Stabil. 88 (2005) 70.
- [14] D. Plusa, M. Dospial, B. Slusarek, U. Kotlarczyk, J. Magn. Magn. Mater. 306 (2006) 302.
- [15] A. Toldy, N. Toth, P. Anna, G. Marosi, Polym. Degrad. Stabil. 91 (2006) 585.
- [16] T. Horikawa, K. Miura, M. Itoh, K. Machida, J. Alloys Compd. 408 (2006) 1386.
- [17] M. Itoh, K. Nishiyama, F. Shogano, T. Murota, K. Yamamoto, M. Sasada, K. Machida, J. Alloys Compd. 451 (2008) 507.
- [18] H.W. Kwon, J.H. Yu, J. Alloys Compd. 487 (2009) 138.
- [19] M. Zakotnik, I.R. Harris, A.J. Williams, J. Alloys Compd. 469 (2009) 314.
- [20] H.W. Chang, M.F. Shih, C.C. Hsieh, W.C. Chang, J. Alloys Compd. 484 (2009) 143.
- [21] S.W.J. Tao, J. Tian, X. Lu, X.H. Qu, Y. Honkura, H. Mitarai, K. Noguchi, J. Alloys Compd. 477 (2009) 510.
- [22] M. Liu, Y. Sun, G.B. Han, W. Yang, R.W. Gao, J. Alloys Compd. 478 (2009) 303.
- [23] M. Liu, G.B. Han, R.W. Gao, J. Alloys Compd. 488 (2009) 310.
- [24] P. Saravanan, R. Gopalan, R. Priya, P. Ghosal, V. Chandrasekaran, J. Alloys Compd. 477 (2009) 322.
- [25] Rong Chuan-bing, Nguyen Van Vuong, J. Liu, J. Ping, Appl. Phys. 107 (2010) 717.
- [26] S.Z. Zhou, Rare-Earth Permanent Magnet Materials and Their Applications, Metallurgy Industry Press, Beijing, 1995, 228.