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Radially anisotropic $Sm_2Fe_{17}N_3/Nd_2Fe_{14}B$ hybrid bonded-magnets using self-organization technique

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Abstract

It has been well accepted that magnetic properties of radially anisotropic ring-shaped bonded-magnets are deteriorated when magnetic field for alignment is limited due to miniaturization. In order to overcome this difficulty, we developed a new technique using self-organization of binder, which enabled the alignment of molecular chain. Furthermore, we achieved the low pressure compacting such as 20 MPa for green compacts by using slip-flow phenomenon. By using these techniques, we made $Sm_2Fe_{17}N_3/Nd_2Fe_{14}B$ rigid void-less sheet magnets with the relative density higher than 98%, controlled their flexibility, transformed them into various shapes, and succeeded in preparing radially anisotropic bonded-magnets. One of examples is a radially anisotropic ring-shaped bonded-magnet which is approximately from 0.35 to 1.0 mm in thickness and less than 15 mm in outer diameter. It could be prepared by rolling a sheet-shaped rigid bonded-magnet with the rolling rate from approximately 3 to 5% at 100–120 °C and curling it. Superior magnetic properties were realized by optimizations of the solidification process of the compound with self-organization of binder under an axially alignment-field and the transformation process into various specific shapes. An obtained $(BH)_{max}$ value exceeded 140 kJ/m^3, even if their thickness is less than 1 mm and the outer diameter is less than 15 mm. Furthermore, the compaction at a low pressure suppresses destruction and cracks of HDDR-$Nd_2Fe_{14}B$ powder, which improved the squareness of the demagnetization curve and the initial flux loss at elevated temperatures such as 100 °C.

Keywords: Rare-earth magnetic powder; Anisotropic rare-earth magnet; Self-organization of binder; Bonded-magnet; Radially anisotropic

1. Introduction

Although usage of anisotropic $Sm_2Fe_{17}N_3$ and $Nd_2Fe_{14}B$ powders is one of promising methods of preparing magnets beyond conventional isotropic $Nd_2Fe_{14}B$ ring-shaped bonded-magnets prepared from melt-spun powder, magnetic properties of radially anisotropic ring-shaped magnets are deteriorated when their outer diameter is decreased. Therefore, radially anisotropic $Sm_2Fe_{17}N_3$ and $Nd_2Fe_{14}B$ bonded-magnets have not been mainly applied to small motors up to now.

In order to overcome the above difficulty, we developed a new technique using self-organization binder, which enabled the alignment of molecular-chain. Previously, by using this technique, we controlled the flexibility of rigid sheet-shaped magnets, transformed them into various shapes, and succeeded in preparing ring- or arc-shaped bonded-magnets [1–5]. Recently, we realized radially anisotropic hybrid-magnets with superior magnetic properties by optimizations of the consolidation process of the self-organization binder under an axially alignment-field and the transformation process into various shapes. In this technique, highly dense green compact can be prepared under a low pressure such as 20 MPa by using the slip-flow phenomenon. Furthermore, we controlled the flexibility of rigid sheet-shaped hybrid-magnets, whose relative density including the binder exceeded 98%, transformed them into various shapes, and prepared radially anisotropic hybrid-magnets. The $(BH)_{max}$ value was from 140 to 160 kJ/m^3, even if their thickness is less than 1 mm and the outer diameter is less than 15 mm. Furthermore, the compacting at a low pressure suppressed destruction and cracks of HDDR-$Nd_2Fe_{14}B$ powder, which improved the

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initial flux loss and the squareness of the demagnetization curve at elevated temperatures such as 100 °C of the prepared hybrid-magnet.

This paper reports our recent achievement on new preparation processes of radially anisotropic hybrid-magnets using the above self-organization technique including slip-flow phenomenon. Magnetic properties of prepared magnets are also reported.

2. Preparation processes

2.1. Materials

HDDR (hydrogenation, disproportionation, desorption, and recombination) $Nd_2Fe_{14}B$ powder (53–150 μm in particle size, 292 kJ/m^3 in $(BH)_{max}$), RD (reduction and diffusion) $Sm_2Fe_{17}N_3$ powder (2–3 μm in particle size, 310 kJ/m^3 in $(BH)_{max}$) were used as starting materials. Solid epoxy-oligomer (70–76 °C in melting point, 205–220 g/eq. in epoxy equivalent), polyamide including an adhesive agent (approximately 80 °C in melting point, 150 μm or less in particle size), pentaerythritol-stearic-tri-ester for slip-agent (51 °C in melting point), and imidazole-adduct (80 °C in melting point, 4 μm or less in particle size) for chemical contact were also used for the self-organization of binder for preparing radially anisotropic $Sm_2Fe_{17}N_3/Nd_2Fe_{14}B$ hybrid-magnets.

2.2. Preparation processes by self-organized binder technique

At first, $Sm_2Fe_{17}N_3$ and $Nd_2Fe_{14}B$ powders were coated with 2 and 0.5 wt.% of solid epoxy-oligomer, respectively. Subsequently, 2.5 wt.% of polyamide including an adhesive agent, 0.25 wt.% of slip-agent, and balance of two kinds of magnetic powder were mixed under a molten state of binder materials. In this process, the particle size of all mixed granular compound was adjusted to 350 μm or less by crushing and sieving-classification. In the final process of the compounding, 0.5 wt.% of polyamide for alignment of molecular chain and 0.3 wt.% of imidazole-adduct for chemical contact were added to the above granular compound.

The above compound was consolidated into a green compact under a pressure from 15 to 50 MPa, an elevated temperature from 150 to 160 °C, and an axially magnetic field of 1.4 MA/m for alignment. The dimensions of the obtained green compact are 25 mm in width, 160 mm in length, and from 0.35 to 1.05 mm in thickness. Then, the prepared green compact was cured at 180 °C for 20 min in air, and a rigid sheet-shaped hybrid-magnet (cured green compact) was obtained. During this process, the imidazole-adduct formed the chemical contact point by cross-linking between epoxy-oligomer and polyamide, and the self-organization phenomenon was completed as shown in Fig. 1.

As the self-organization of binder enables the alignment of molecular-chain, we can control the flexibility of hybrid-magnets, transform them into various shapes. For the prepared hybrid-magnets, the compactibility, flexibility after the alignment of molecular-chain, and magnetic properties were evaluated.

3. Results and discussion

3.1. Fabrication of hybrid-magnet by using self-organization technique

The density of a magnet is shown in Fig. 2 as a function of the compacting pressure for two kinds of gran-

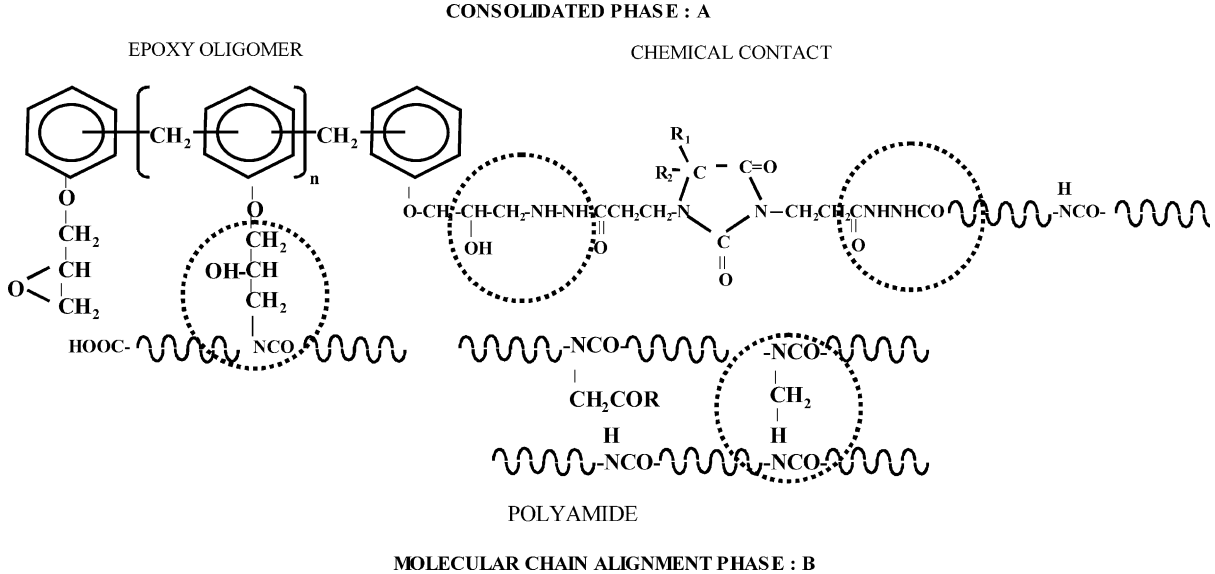

Fig. 1. Chemical structure of self-organized binder prepared by reaction of "Phase-A", "Phase-B", and their chemical contact point. In the figure, the dotted circles indicate the estimated chemical structures of the chemical contacts points.

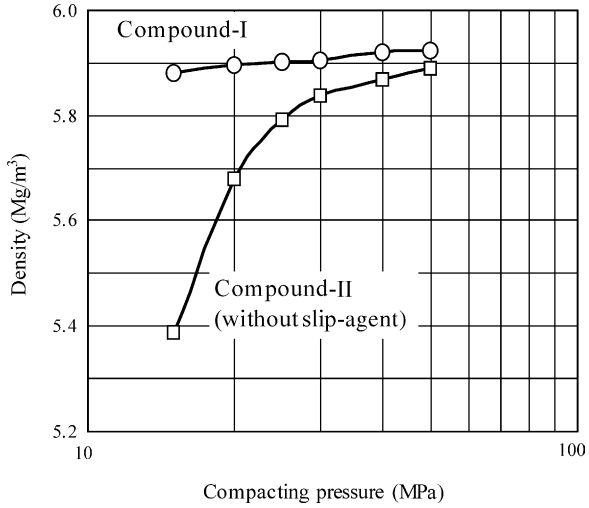

Fig. 2. Density of magnet as a function of compacting pressure. The results denoted by Compound-I, and -II indicate those for the mixture in molten state with slip-agent, and the mixture in molten state without slip-agent.

ular compound, Compound-I, and -II. For Compound-II, highly dense magnet was obtained under a pressure exceeding 50 MPa as shown in Fig. 2. On the other hand, a high density was achieved under the pressure of 20 MPa for Compound-I. This pressure is approximately 1/30 or less compared with that needed for the conventional isotropic $Nd_2Fe_{14}B$-based solid-epoxy compound, typically from 600 to 1000 MPa. A small pressure required for Compound-I can be attributed to the fact that the self-organized binder exhibits the slip-flow phenomenon. This slip-flow phenomenon was seen significantly for the optimized compound, Compound-I, at 150–160 °C. In addition, slip-agent reduces the friction among the self-organization of binder, the magnetic powder, and compaction-dies. The density and relative density including binder of magnets prepared from Compound-I exceeded 5.9 Mg/m^3 and 98%, respectively, under a low pressure such as 20 MPa, when the self-organization binder concentration was 3.3 wt.%. Thus, we can conclude that Compound-I exhibits good compactibility with assistance of the slip-flow phenomenon.

A SEM micrograph of the cross-section of a 350 μm-thick hybrid-magnet is shown in Fig. 3. The observed magnet is the thinnest one in the magnets obtained in this study and the weight ratio of $Sm_2Fe_{17}N_3$ and $Nd_2Fe_{14}B$ including in the magnet is 40/60. As seen in the figure, two kinds of powder are uniformly distributed even if their thickness is 350 μm, and good compactibility is observed, suggesting that $Nd_2Fe_{14}B$ was integrated together with $Sm_2Fe_{17}N_3$ with a good hybridization. Furthermore, no cracks were observed in the $Nd_2Fe_{14}B$ powder. Therefore, the developed techniques have an advantage in preparation process. Namely, the low pressure compacting suppresses destruction and cracks of $Nd_2Fe_{14}B$ powder, which is expected to improve the reliability of the magnets as discussed below.

Fig. 4 shows the changes in the distribution of particle size for Compound-I. As seen in the figure, the distribution did not change before and after the compaction. It is known that magnetic properties of magnets are degraded by the destruction and damage of the $Nd_2Fe_{14}B$ powder due to compaction. The unchanged distribution suggests that the usage of our self-organizing technique with slip-flow phenomenon is expected to suppress the above-mentioned degradation.

Furthermore, the comparison of the results of particle size distribution curves due to compaction as shown in Fig. 4 clearly shows the buffer effect of containing $Sm_2Fe_{17}N_3$ powder.

3.2. Control of flexibility and magnetic properties

The control of the flexibility is an important item in transformation of the prepared rigid sheet-shaped magnet into the radially anisotropic ring-shaped magnet. An effective method of giving the flexibility to the prepared hybrid-magnet is usage of the hot rolling. One of examples is a radially

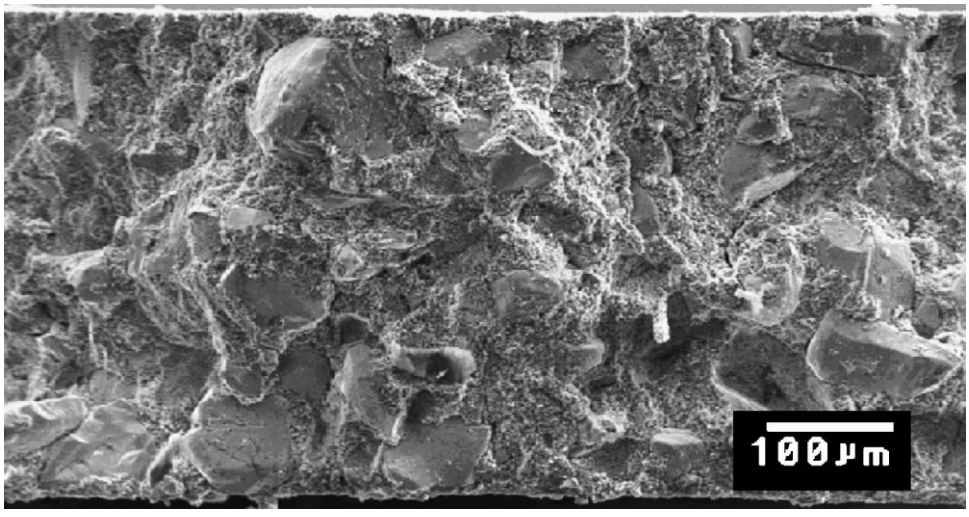

Fig. 3. SEM micrograph of cross-section of the thinnest hybrid-magnet prepared by using slip-flow phenomenon.

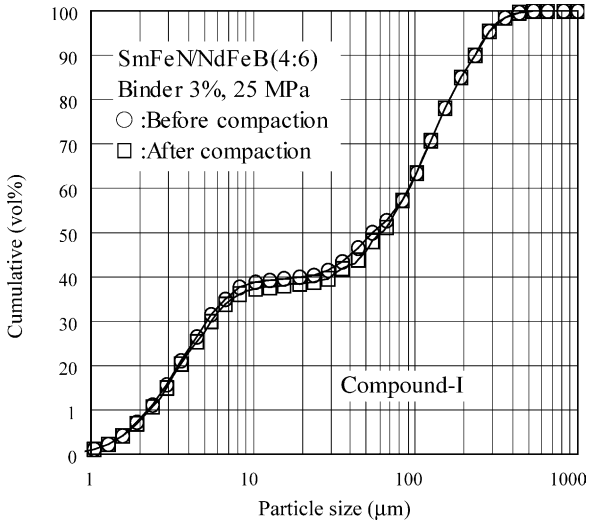

Fig. 4. Changes in distribution of particle size for Compound-I. The distributions of particle size were measured with a laser-diffraction-particle-analyzer after the solvent extraction of the samples.

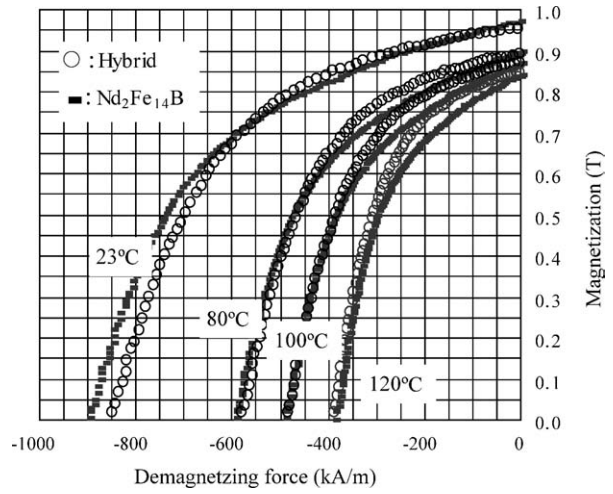

Fig. 6. Demagnetization curves of the hybrid-magnet, together with $Nd_2Fe_{14}B$-based bonded-magnet. The demagnetization curves were measured with a VSM after magnetization by a pulsed field of 4 MA/m.

anisotropic ring-shaped bonded-magnet which is approximately from 0.35 to 1.0 mm in thickness and from 6 to 15 mm in outer diameter. It could be prepared by rolling a sheet-shaped rigid hybrid-magnet with the rolling rate approximately from 3 to 5% and curling it at room temperature as shown in Fig. 5. The $(BH)_{max}$ value exceeded 140 kJ/m^3, even if their thickness is less than 1 mm and the outer diameter is less than 15 mm.

Fig. 6 shows typical demagnetization curves of the hybrid-magnet prepared from Compound-I, together with those for anisotropic $Nd_2Fe_{14}B$ bonded-magnet. The dimensions and density of the test specimen are 8 mm in width, 3.5 mm in thickness, 8 mm in length, and 5.91 Mg/m^3 in density, respectively. The remanence values J_r and the magnetization values at the working point of $B/\mu_0 H = 1$ were almost the same for both the magnets, and their $(BH)_{max}$ values exceeded 160 kJ/m^3. The difference between two kinds of magnets was clearly observed in the squareness, H_k/H_{CJ}, of the demagnetization curve at the elevated temperatures. Here, H_k and H_{CJ} are a demagnetizing force corresponding to the magnetization of $0.9J_r$ and intrinsic coercivity, respectively.

Previously, we systematically evaluated the flux loss in bonded-magnets with various values of coercivity, H_{CJ}, and found that initial flux losses in some nanocomposite rare-earth bonded-magnets were smaller than that of a conventional isotropic $Nd_2Fe_{14}B$ bonded-magnet, despite their small coercivity, H_{CJ} values [6,7]. We also clarified that the observed small flux loss can be attributed to an improvement in the squareness, H_k/H_{CJ}, of the demagnetization curves at an elevated temperature for the nanocomposite magnets studied. Therefore, the improvement in the initial flux loss seen in Fig. 7 is expected to be attributed to the squareness, H_k/H_{CJ}, at 100 °C of the hybrid-magnets.

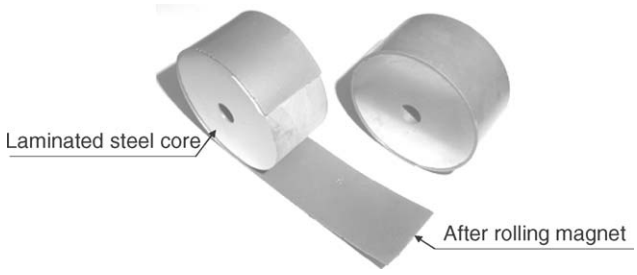

Fig. 5. External views of a radially anisotropic ring-shaped hybrid-magnets. The intended dimensions of the magnets are 50 mm in outer diameter and 1.05 mm in thickness.

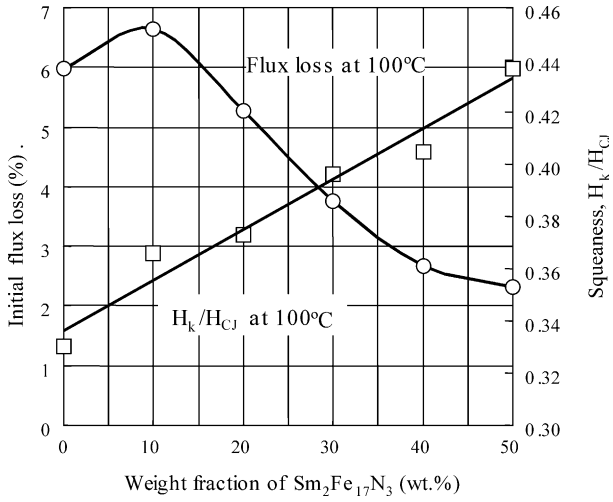

Fig. 7. Initial flux loss of a hybrid-magnet, and squareness, H_k/H_{CJ}, at 100 °C as a function of the fraction of $Sm_2Fe_{17}N_3$. The coercivity H_{CJ} was almost the same (470–480 kA/m) at 100 °C. The coercivity and squareness were measured with a VSM after magnetization by a pulsed field of 4 MA/m.

4. Conclusion

We have developed a new technique, which enabled us to prepare a radially anisotropic $Sm_2Fe_{17}N_3/Nd_2Fe_{14}B$ hybrid-magnets, 15 mm or less in outer diameter and 350 μm or more in thickness, by using self-organization technique including slip-flow phenomenon for preparation processes. The $(BH)_{max}$ value exceeded 140 kJ/m^3, even if their thickness is less than 1 mm and the outer diameter is less than 15 mm. Furthermore, they can be made without destruction and cracks of HDDR-$Nd_2Fe_{14}B$ powder, which improved the squareness, H_k/H_{CJ}, of the demagnetization curve and the initial flux loss at elevated temperatures such as 100 °C.

References

[1] F. Yamashita, Proceedings of the 17th International Workshop on Rare-earth Magnets and their Applications, Newark, 2002, p. 100.
[2] F. Yamashita, A. Watanabe, H. Fukunaga, IEE Trans. FM 123 (2003) 146.
[3] F. Yamashita, A. Watanabe, H. Fukunaga, IEEE Trans. Magn. 39 (2003) 2980.
[4] F. Yamashita, A. Watanabe, S. Tsutsumi, H. Fukunaga, J. Magn. Magn. Mater 272–276 (2004) 1865.
[5] F. Yamashita, S. Tsutsumi, H. Fukunaga, Ninth Joint MMM/INTERMAG, CA, 2004, BP-05.
[6] H. Fukunaga, S. Hayashida, Y. Kanai, F. Yamashita, J. Magn. Magn. Mater 203 (1999) 304.
[7] Y. Kanai, S. Hayashida, H. Fukunaga, F. Yamashita, IEEE Trans. Magn. 35 (1999) 3292.