Journal of Magnetism and Magnetic Materials 467 (2018) 64–68

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Research articles

Increase of energy products of Zn-bonded Sm-Fe-N magnets with low oxygen content

Masashi Matsuura[a,*], Yuki Nishijima[a], Nobuki Tezuka[a], Satoshi Sugimoto[a], Tetsuya Shoji[b,c], Noritsugu Sakuma[b,c]

[a] Department of Materials Science, Graduate School of Engineering, Tohoku University, 6-6-02 Aoba-yama, Sendai, Miyagi 980-8579, Japan
[b] Toyota Motor Corporation, 1 Toyota-cho, Toyota, Aichi 471-8572, Japan
[c] Technology Research Association of Magnetic Materials for High-efficiency Motors, (MagHEM) Higashifuji-Branch, 1200 Mishuku, Susono, Shizuoka 410-1193, Japan

ARTICLE INFO

Keywords:
Sm-Fe-N magnets
Zn binder
Binder less
Maximum energy products
Oxygen content
Arc plasma deposition
Spark plasma sintering

ABSTRACT

To increase the maximum energy product, $(BH)_{max}$, of Zn-bonded Sm-Fe-N magnets, a process for preparing a Sm-Fe-N powder with a low oxygen content was developed. The Sm-Fe-N powder containing 0.23 wt% oxygen was prepared by ball milling and nitriding, and the powder remanence and coercivity were 140 $A{\cdot}m^2kg^{-1}$ and 0.72 $MA{\cdot}m^{-1}$, respectively.

Arc plasma deposition (APD) was used for Zn addition. Fine Zn particles several tens of nanometers in size were deposited on the Sm-Fe-N powder under vacuum conditions, and APD prevented the oxygen content increasing in the Zn-deposited Sm-Fe-N powder. The low-oxygen Sm-Fe-N powder was pressed and sintered by spark plasma sintering (SPS), and 3.3 wt% Zn-bonded and Zn-free Sm-Fe-N magnets were prepared. For both Zn-bonded and Zn-free Sm-Fe-N magnets, the density increased with increasing sintering pressure, and the relative density of the Zn-bonded Sm-Fe-N magnet was 89% and that of the Zn-free Sm-Fe-N magnet was 85% after SPS. The coercivities of the Zn-bonded and Zn-free Sm-Fe-N magnets were both high at 1.1 and 0.8 $MA{\cdot}m^{-1}$, respectively. $(BH)_{max}$ of these magnets increased with increasing relative density, and the highest $(BH)_{max}$ value of the Zn-bonded Sm-Fe-N magnets was 153 $kJ{\cdot}m^{-3}$ and that of the Zn-free Sm-Fe-N magnet was 179 $kJ{\cdot}m^{-3}$.

1. Introduction

$Sm_2Fe_{17}N_3$ has a high saturation magnetic polarization (J_s), a large anisotropy field, and a high Curie temperature [1,2], however, $Sm_2Fe_{17}N_3$ cannot be sintered because it decomposes above about 600 °C. Therefore, $Sm_2Fe_{17}N_3$ based Sm-Fe-N powders are used in bonded magnets. Resin-bonded $Sm_2Fe_{17}N_3$ magnets are used in several motors. Metal-bonded Sm-Fe-N magnets, which have a metal with a low melting point as a binder, have been studied extensively. Otani et al. [3] investigated the magnetic properties of metal-bonded $Sm_2Fe_{17}N_3$ magnets. They reported that the Zn binder improved the coercivity of the Sm-Fe-N bonded magnets, and the appearance of Zn_7Fe_3, which is the Γ-FeZn phase, increased the coercivity. To increase $(BH)_{max}$ of the Zn-bonded Sm-Fe-N magnets, it is necessary to increase the relative density and volume fraction of Sm-Fe-N powder.

Ito et al. [4] reported that hot isostatic pressing can improve the relative density of Zn-bonded Sm-Fe-N magnets, and they reported that $(BH)_{max}$ of the 5 wt% Zn-bonded Sm-Fe-N magnet was 168 $kJ{\cdot}m^{-3}$. However, the coercivity of the magnet was still low at 564 $kA{\cdot}m^{-1}$. Machida and co-workers [5,6] have reported several Zn-bonded Sm-Fe-N magnets. They used Zn and In-Zn as binders and prepared bonded magnets by pressing at 1.4 GPa. The density, $(BH)_{max}$, and coercivity were 6.5 $kg{\cdot}m^{-3}$, approximately 151 $kJ{\cdot}m^{-3}$, and 0.72 $MA{\cdot}m^{-1}$, respectively. Saito et al. [7] and Prabhu et al. [8] prepared high-density Zn-bonded Sm-Fe-N magnets by spark plasma sintering (SPS). Saito et al. [7] prepared 5 and 10 wt% Zn-bonded Sm-Fe-N magnets by magnetic-field-assisted SPS, and the magnets had relative densities of 88.7–92.5% and $(BH)_{max}$ of 158 $kJ{\cdot}m^{-3}$. Prabhu et al. [8] reported a high coercivity of 2.19 $MA{\cdot}m^{-1}$, a high density of 6.2 $kg{\cdot}m^{-3}$, and a remanence of 0.62 T in the 15 wt% Zn-bonded Sm-Fe-N magnet. Takagi et al. [9] and Soda et al. [10] reported binder-less Sm-Fe-N magnets prepared by SPS. Soda et al. [10] prepared Sm-Fe-N powder with a low oxygen content, and they applied a high sintering pressure of 1.2 GPa to fabricate binder-less Sm-Fe-N magnets with densities of 86–88% and $(BH)_{max}$ of about 160 $kJ{\cdot}m^{-3}$. They also showed that the coercivity of low-oxygen Sm-Fe-N magnets can be kept the same as that of the raw powders. Oxygen at the surface of the Sm-Fe-N powder can decrease the coercivity [9,11] because the oxygen decomposes the $Sm_2Fe_{17}N_3$ phase.

* Corresponding author.
E-mail address: m-matsu@material.tohoku.ac.jp (M. Matsuura).

https://doi.org/10.1016/j.jmmm.2018.07.064
Received 17 March 2018; Received in revised form 7 June 2018; Accepted 20 July 2018
Available online 21 July 2018

Therefore, reducing the oxygen content in the Sm-Fe-N powder suppresses the decrease in coercivity.

We reported high-coercivity Zn-bonded Sm-Fe-N magnets [12] prepared by decreasing the oxygen content and particle size of the Zn powder. The 15 wt% Zn-bonded Sm-Fe-N magnets showed the high coercivity of 2.66 $MA{\cdot}m^{-1}$, and the 10 wt% Zn-bonded magnet had a high coercivity value of 2.41 $MA{\cdot}m^{-1}$ and an energy product of 56.1 $kJ{\cdot}m^{-3}$. We have also reported high-density Zn-bonded Sm-Fe-N magnets fabricated by mechanical processing. Ishihara et al. [13] produced 5 wt% Zn-bonded Sm-Fe-N magnets with a high relative density of 97%, high $(BH)_{max}$ of 132 $kJ{\cdot}m^{-3}$, and coercivity of 1.36 $MA{\cdot}m^{-1}$ by hot-rolling. Kataoka et al. [14] prepared 15 wt% Zn-bonded Sm-Fe-N magnets by swaging with a relative density of about 90%, $(BH)_{max}$ of 67.8 $kJ{\cdot}m^{-3}$, and coercivity of 1.89 $MA{\cdot}m^{-1}$.

These studies suggest that increasing the relative density and decreasing the oxygen content in Zn-bonded Sm-Fe-N magnets increases $(BH)_{max}$ while maintaining a high coercivity. In addition, decreasing the particle size of Zn and increasing the homogeneity of the Zn distribution are also important for increasing the coercivity. In other words, fine, homogeneously distributed Zn can increase the coercivity of Zn-bonded Sm-Fe-N magnets and decrease the Zn content, leading to an increase in the volume fraction of Sm-Fe-N powder and an increase in $(BH)_{max}$.

To achieve the low oxygen content, high density, and small amount of Zn required to produce high-performance Sm-Fe-N magnets, we focused on arc plasma deposition (APD) for coating Sm-Fe-N powder with Zn and SPS for increasing the density of Zn-bonded Sm-Fe-N magnets. In APD, the target is vaporized and ionized by the arc discharge, and fine particles are deposited on the substrate or particles. The nanoparticle morphology and amount of deposition can be controlled by the deposition conditions, such as deposition pressure, atmosphere, discharge voltage, and discharge count. APD can deposit fine Zn particles with a low oxygen content on Sm-Fe-N powder [15,16] because it is performed under high-vacuum conditions. To decrease the oxygen content of Zn-bonded Sm-Fe-N magnets further, Sm-Fe-N powder with a low oxygen content is also needed. SPS uses a pulsed DC current to heat materials rapidly under uniaxial pressure. SPS can produce high-density compacts at a low sintering temperature and short sintering time.

In this study, we increase $(BH)_{max}$ of Sm-Fe-N magnets. Fine Zn particles were deposited on Sm-Fe-N powder by APD under high-vacuum conditions, and the relationship between morphology and coercivity was investigated. Low oxygen content Sm-Fe-N powder was fabricated, and Zn was deposited on the Sm-Fe-N powder. High-density Sm-Fe-N magnets were prepared from the low-oxygen powder by SPS.

2. Experimental procedure

Fig. 1 shows a flow diagram of the sample preparation. We used commercial Sm-Fe-N powder (average particle size ≈3 μm) and the other is low oxygen content Sm-Fe-N powder ("Low-O Sm-Fe-N"), which was prepared from Sm_2Fe_{17}-based Sm-Fe powder. Sm-Fe powder, which was obtained by reduction diffusion process, was pulverized by ball milling at a rotation speed of 150 rpm and milling time of 30 min in organic solvent. The fine Sm-Fe powder was dried in an inert gas glove box and was nitrided under N_2 gas at 450 °C for 10 h.

Zn was deposited by APD with a base pressure of under 1×10^{-4} Pa. Commercial or Low-O Sm-Fe-N powder was put in a cup and placed in the APD chamber. Zn was deposited on the Sm-Fe-N powder with a discharge voltage of 150 V and discharge count of 12,000–45,000. By controlling discharge counts of APD, amount of Zn can be controlled. During Zn deposition, the Sm-Fe-N powder was agitated intermittently in APD chamber. For comparison, commercial Zn powder with average particle sizes of < 7 μm was also used. The commercial Zn and commercial Sm-Fe-N powders were mixed by ball milling with a rotation speed of 150 rpm for 30 min in organic solvent for preventing oxidation. The Sm-Fe-N powders containing either

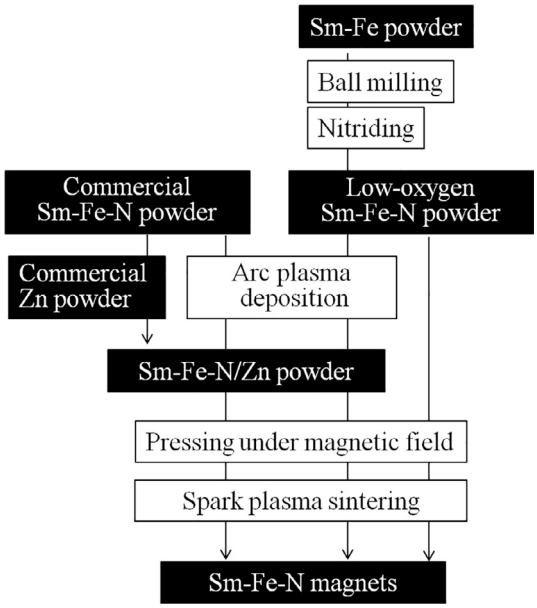

Fig. 1. Flow diagram of sample preparation.

deposited or mixed Zn particles ("Sm-Fe-N/Zn" powder) were pressed at 200 MPa under an applied magnetic field of 2.3 $MA{\cdot}m^{-1}$ in an inert gas atmosphere. The green compact was sintered by SPS under a sintering pressure of 400, 600, or 750 MPa, a sintering temperature of 420–460 °C, and under vacuum. A Zn-free Sm-Fe-N magnet was also prepared from Low-O Sm-Fe-N powder by SPS.

The magnetic properties of the Sm-Fe-N magnets were measured by a DC B-H loop tracer. Oxygen content was measured by an O/N analyzer and the microstructure was observed by field-emission scanning electron microscopy (SEM). The powder composition was measured by X-ray fluorescence (XRF).

3. Results and discussion

3.1. Increased dispersibility of fine, low-oxygen Zn particles by APD

A Zn binder can increase the coercivity of Zn-bonded Sm-Fe-N magnets, although the Zn content must be decreased to increase $(BH)_{max}$. Therefore, APD was used to deposit fine, well-dispersed Zn with a low oxygen content on Sm-Fe-N powder. Fig. 2 shows SEM and Zn-mapping images of the commercial Sm-Fe-N/Zn powder obtained by APD. The Zn discharge count was 20,000 shots, and XRF measurements

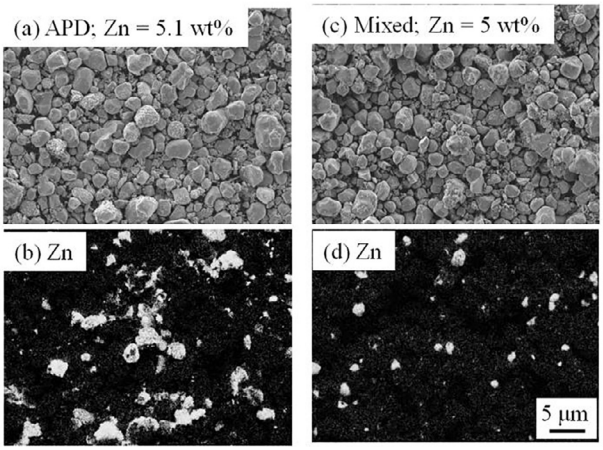

Fig. 2. SEM and Zn-mapping images of Sm-Fe-N/Zn powders. (a)(b) APD and (c)(d) mixed Sm-Fe-N/Zn powder.