



# Article Production of Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> Bulk Magnets

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Abstract: Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder exhibits excellent magnetic properties but is unstable and decomposes into  $\alpha$ -Fe and SmN phases at high temperatures. Therefore, the key to producing Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets is to reduce the deterioration of Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder during sintering. Herein, Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets were made using the spark plasma sintering (SPS) method with the addition of zinc stearate powder and zinc powder. Adding small amounts of zinc stearate powder and zinc powder improved the magnetic anisotropy and the coercivity of the magnets, respectively. The magnets produced by the SPS method using zinc stearate powder and zinc powder exhibited enhanced magnetic properties almost comparable to those of Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder.

Keywords: permanent magnet; rare-earth compound; spark plasma sintering; magnetic property

### 1. Introduction

High-performance neodymium-iron-boron (Nd-Fe-B) magnets have been applied to various advanced electromagnetic devices, including hard disk drives, electric vehicles, and medical equipment [1–3]. In particular, the production of electric cars equipped with Nd-Fe-B magnet motors has considerably increased owing to the global trend of enacting regulations to reduce the production of internal combustion engines, which emit large volumes of greenhouse gases [4]. The growing demand for high-performance Nd-Fe-B magnets has raised concerns over the prices of rare-earth materials and their availability [5–10]. Therefore, the development of permanent magnets using the relatively abundant rare-earth element Sm has recently attracted interest.

Samarium-iron-nitride ( $Sm_2Fe_{17}N_3$ ) powder consists of the  $Sm_2Fe_{17}N_3$  compound. The  $Sm_2Fe_{17}N_3$  compound has been produced by the nitrogenation of  $Sm_2Fe_{17}$  alloy powder, and thus, the  $Sm_2Fe_{17}N_3$  compound has been made in powder form. The  $Sm_2Fe_{17}N_3$ compound possesses not only high saturation magnetization comparable to the  $Nd_2Fe_{14}B$ compound but also a higher Curie temperature and a higher uniaxial anisotropy [11–13]. However, the phase decomposition of the  $Sm_2Fe_{17}N_3$  compound at high temperatures above 873 K makes it difficult to produce  $Sm_2Fe_{17}N_3$  bulk magnets in practice [14]. Several attempts have been made to integrate  $Sm_2Fe_{17}N_3$  powder into bulk magnets [15–18].  $Sm_2Fe_{17}N_3$  bulk magnets have been produced by nonconventional consolidation techniques such as dynamic compaction using explosion or gun methods [15,16]. The dynamic compaction has realized high-density  $Sm_2Fe_{17}N_3$  bulk magnets. Another nonconventional consolidation technique, the so-called compression shearing method, has been successfully applied to produce the  $Sm_2Fe_{17}N_3$  bulk magnets [17]. The compression shearing method has also realized high-density  $Sm_2Fe_{17}N_3$  bulk magnets. However, there has yet to be a practical application of these techniques due to the difficulty in production.

The SPS method has recently been effectively applied to produce  $Sm_2Fe_{17}N_3$  bulk magnets [18,19]. The addition of zinc powder has been suggested to improve the coercivity of  $Sm_2Fe_{17}N_3$  bulk magnets [20,21]. However, these magnets were magnetically isotropic, and their remanence magnetization was not comparable to that of  $Sm_2Fe_{17}N_3$  powder. The key to producing high-performance  $Sm_2Fe_{17}N_3$  bulk magnets using the SPS method is to increase their remanence. As  $Sm_2Fe_{17}N_3$  powder possesses high magnetization, one



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**Copyright:** © 2024 by the author. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). solution is to align  $Sm_2Fe_{17}N_3$  powder before sintering. Herein, anisotropic  $Sm_2Fe_{17}N_3$  bulk magnets were successfully produced using the SPS method with the addition of zinc stearate powder and zinc powder, and the effects of their addition on the magnetic properties of the magnets were examined.

#### 2. Results and Discussion

The images of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder and Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder are shown in Figure 1. The Sm<sub>2</sub>Fe<sub>17</sub> alloy powder was made using the diffusion reduction process [22]. The Sm<sub>2</sub>Fe<sub>17</sub> alloy powder was a coarse powder with an average particle size of about 30  $\mu$ m. The Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder was first made by nitrogenation of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder and then pulverized into fine powder. The average particle size of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder was about 3  $\mu$ m. The fine Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder was used in this study.



Figure 1. SEM images of (a) Sm<sub>2</sub>Fe<sub>17</sub> alloy powder and (b) Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder.

Before the consolidation of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder using the SPS method, the powder was characterized by X-ray diffraction and thermomagnetic studies. Figure 2 shows the X-ray diffraction pattern of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder, together with that of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder. The X-ray diffraction pattern of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder shows a typical Th<sub>2</sub>Zn<sub>17</sub>-type phase. The diffraction peaks of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder were indexed to be the Th<sub>2</sub>Zn<sub>17</sub>-type phase, and no clear diffraction peaks of Sm-oxides and  $\alpha$ -Fe phase were detected in the X-ray diffraction pattern. This indicates that the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder made using the diffusion reduction process consisted of the Th<sub>2</sub>Zn<sub>17</sub>-type Sm<sub>2</sub>Fe<sub>17</sub> phase. The X-ray diffraction pattern of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder also exhibits a typical Th<sub>2</sub>Zn<sub>17</sub>-type phase. Although it is known that the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase decomposes into the  $\alpha$ -Fe and SmN phases at high temperatures [14], no diffraction peaks of these phases were noticed in the X-ray diffraction pattern. It is confirmed that the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder consisted of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase.

Figure 3 shows the thermomagnetic curve of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder, together with that of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder. It was found that the thermomagnetic curves of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder and the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder are pretty different. The thermomagnetic curve of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder shows a magnetic transition at around 490 K. According to the results of the X-ray diffraction studies, the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder consisted of the Th<sub>2</sub>Zn<sub>17</sub>-type Sm<sub>2</sub>Fe<sub>17</sub> phase. Thus, the magnetic transition is believed to be the Curie temperature of the Sm<sub>2</sub>Fe<sub>17</sub> phase. On the other hand, a large magnetic transition at around 740 K is observed in the thermomagnetic curve of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder. Since the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder consisted of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase, the magnetic transition is believed to be the Curie temperature of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase. The Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase has a significantly high Curie temperature of 740 K, compared with the Nd<sub>2</sub>Fe<sub>14</sub>B phase in the Nd-Fe-B magnets (585 K) [23]. The observed Curie temperature of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase is consistent with the reported value [11]. No magnetic transition of the Sm<sub>2</sub>Fe<sub>17</sub> phase was noted in the thermomagnetic curve, indicating that the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder solely consisted of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase.



**Figure 2.** X-ray diffraction patterns of (**a**)  $\text{Sm}_2\text{Fe}_{17}$  alloy powder and (**b**)  $\text{Sm}_2\text{Fe}_{17}\text{N}_3$  powder. These diffraction peaks were indexed by the PDF cards (#00-048-1789 for  $\text{Sm}_2\text{Fe}_{17}$  and #01-079-9758 for  $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ ).



**Figure 3.** Thermomagnetic curves of (**a**)  $Sm_2Fe_{17}$  alloy powder and (**b**)  $Sm_2Fe_{17}N_3$  powder. The arrows indicate the magnetic transition temperature.

However, a slight increase in magnetization at temperatures over the Curie temperature is observed in the thermomagnetic curve of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder. This suggests that a new magnetic phase formed during the thermomagnetic measurement. The new magnetic phase is believed to be the  $\alpha$ -Fe phase since the new magnetic phase shows the Curie temperature near 1050 K. The thermomagnetic study indicates that the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase decomposes into the  $\alpha$ -Fe and SmN phases at high temperatures over 800 K. The observed decomposition temperature is slightly lower than the reported value (873 K) [14]. In any case, it is essential to sinter the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder at the lowest possible temperature because the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase in Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder decomposes into the  $\alpha$ -Fe and SmN phases at high temperatures.

In order to confirm the decomposition of the  $Sm_2Fe_{17}N_3$  powder, the specimen after the thermomagnetic measurement was examined using X-ray diffraction. The result is shown in Figure 4. No diffraction peaks of the  $Sm_2Fe_{17}N_3$  phase were seen in the X-ray diffraction pattern. Instead, diffraction peaks of the  $\alpha$ -Fe and SmN phases were noticed in the X-ray diffraction pattern. This confirms that the  $Sm_2Fe_{17}N_3$  phase decomposes into the  $\alpha$ -Fe and SmN phases during thermomagnetic measurement.



**Figure 4.** X-ray diffraction patterns of (**a**)  $Sm_2Fe_{17}N_3$  powder and (**b**) the specimen after the thermomagnetic measurement.

Figure 5 shows the hysteresis loops of the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder and the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder. The Sm<sub>2</sub>Fe<sub>17</sub> alloy powder showed a narrow hysteresis loop, but the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder exhibited a wide one. This indicates that the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder without nitrogenation did not display large coercivity. On the other hand, Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder, prepared by nitriding the Sm<sub>2</sub>Fe<sub>17</sub> alloy powder, exhibited a large coercivity. The Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder exhibits a large coercivity of 12.4 kOe with a remanence of 75.3 emu/g.



Figure 5. Hysteresis loops of (a) Sm<sub>2</sub>Fe<sub>17</sub> alloy powder and (b) Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder.

In order to confirm the magnetic anisotropy of the  $Sm_2Fe_{17}N_3$  powder, the magnetic properties of the  $Sm_2Fe_{17}N_3$  powder were measured parallel to the magnetic alignment after the  $Sm_2Fe_{17}N_3$  powder had been magnetically aligned. Figure 6 shows the hysteresis loops of the  $Sm_2Fe_{17}N_3$  powder measured parallel and perpendicular to the direction of the magnetic alignment. The respective hysteresis loops are quite different. This confirms that the  $Sm_2Fe_{17}N_3$  powder is magnetically anisotropic. The powder exhibited a high remanence of 107 emu/g and a large coercivity of 10.3 kOe when measured parallel to the direction of the magnetic alignment.



**Figure 6.** Hysteresis loops of the  $Sm_2Fe_{17}N_3$  powder, measured parallel (//) and perpendicular ( $\perp$ ) to the direction of the magnetic alignment.

In this study, the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder was sintered into bulk magnets carried out at 673 K, far below its decomposition temperature of 800 K. Since the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder is magnetically anisotropic, the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder was magnetically aligned before sintering (see Figure 7). Despite the low sintering temperature, Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets were produced from the magnetically aligned Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder using the SPS method.

Carbon contamination from carbon dies might occur in the SPS method [24]. To avoid this, the SPS experiment was done with a BN-coated carbon die, and the surface of the  $Sm_2Fe_{17}N_3$  bulk magnets was ground to remove carbon contamination before the property measurements.



**Figure 7.** Schematic diagram showing the setup for the magnetic alignment. Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder was poured into a carbon die and then aligned under an applied magnetic field.

A photograph of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnet is shown in Figure 8. No significant cracks were found in the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnet. The relative density of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnet was as high as 91.7% (compared with the density of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder). A higher consolidation temperature is known to promote the densification of bulk materials produced using the SPS method [25]. A further increase in the consolidation temperature

may result in an increase in density, but the consolidation temperature was kept below 673 K in this study because the  $Sm_2Fe_{17}N_3$  phase in  $Sm_2Fe_{17}N_3$  powder easily decomposes into the  $\alpha$ -Fe and SmN phases at high temperatures.



**Figure 8.** A photograph of the  $Sm_2Fe_{17}N_3$  bulk magnet obtained from the  $Sm_2Fe_{17}N_3$  powder using the SPS method. The magnet was prepared at 673 K under an applied pressure of 100 MPa for 300 s using the SPS method. The magnet had a density of 7.03 g/cm<sup>3</sup> (approximately 91.7% of the powder).

Figure 9 shows the magnetic hysteresis loops of the  $Sm_2Fe_{17}N_3$  powder and the magnet produced. The powder exhibited a high remanence of 107 emu/g and a large coercivity of 10.3 kOe. However, the magnets showed a remanence of 68.6 emu/g and a coercivity of 3.05 kOe. This indicates that the magnetic properties of the powder deteriorated during sintering. Thus, it is essential to reduce the deterioration of the magnetic properties of  $Sm_2Fe_{17}N_3$  powder during sintering.



**Figure 9.** Hysteresis loops of the magnetically aligned  $Sm_2Fe_{17}N_3$  powder and  $Sm_2Fe_{17}N_3$  bulk magnet, prepared at 673 K under an applied pressure of 100 MPa for 300 s using the SPS method.

As remanence is related to the magnetic alignment of the powder in the magnet, small amounts of sintering aid powder were added to the  $Sm_2Fe_{17}N_3$  powder before magnetic alignment, and its effects on the magnetic properties of the magnets were examined. In this study, various sintering aid powders, such as graphite and silica powders, were applied for this purpose. It was found that the zinc stearate powder could improve the density and magnetic alignment of the  $Sm_2Fe_{17}N_3$  powder. The effects of zinc stearate powder addition

on the relative density of the  $Sm_2Fe_{17}N_3$  bulk magnets are shown in Figure 10. The relative density of the  $Sm_2Fe_{17}N_3$  bulk magnet increased from 91.7% to 95.8% as the zinc stearate content increased. Although the density of the zinc stearate powder was 1.095 g/cm<sup>3</sup>, much smaller than that of the  $Sm_2Fe_{17}N_3$  powder (7.66 g/cm<sup>3</sup>), the zinc stearate powder drastically improved the density of the  $Sm_2Fe_{17}N_3$  bulk magnet. The resultant  $Sm_2Fe_{17}N_3$  bulk magnet with small amounts of zinc stearate powder had a high density of over 95%.



Figure 10. Dependence of the relative density of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets on the zinc stearate content.

Figure 11 shows the effects of zinc stearate powder addition on the remanence and coercivity of the magnets. The remanence significantly increased but then decreased as the amount of zinc stearate powder was increased. In contrast, the coercivity gradually decreased as the amount of zinc stearate increased. This confirms that the addition of zinc stearate powder before magnetic alignment improved the magnetic alignment of the  $Sm_2Fe_{17}N_3$  powder.



**Figure 11.** Dependence of the remanence and coercivity of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets on the zinc stearate content.

Figure 12 shows the hysteresis loops of the  $Sm_2Fe_{17}N_3$  bulk magnet with 1 wt% zinc stearate powder measured parallel and perpendicular to the magnetic alignment in order to examine the magnetic anisotropy. The  $Sm_2Fe_{17}N_3$  bulk magnet is anisotropic and shows a higher remanence, 90.6 emu/g, in the parallel direction than the perpendicular direction. The magnet exhibits magnetic anisotropy due to the magnetic alignment of the  $Sm_2Fe_{17}N_3$  powder.



**Figure 12.** Hysteresis loops of the  $Sm_2Fe_{17}N_3$  bulk magnets with 1 wt% zinc stearate powder measured parallel (//) and perpendicular ( $\perp$ ) to the direction of the magnetic alignment. In the hysteresis measurements, rod-like specimens (1 × 1 × 10 mm) were cut from the magnets to avoid demagnetization correction.

As the magnetic alignment of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder is determined via the crystallographic alignment in the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase, the crystallographic alignment of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase in the magnets was examined using X-ray diffraction patterns. Figure 13 shows the X-ray diffraction patterns of the magnet with 1% zinc stearate powder. As shown in the figure, X-ray diffraction patterns were measured perpendicularly and parallel to the direction of magnetic alignment. The X-ray diffraction pattern of the magnet shows a typical powder pattern of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase when measured perpendicular ( $\perp$ ) to the direction of magnetic alignment. However, when measured parallel (//) to the direction of magnetic alignment, the X-ray diffraction pattern exhibited a prominent (006) peak of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase owing to the crystallographic alignment of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase in the magnet. This confirms that the crystallographic alignment of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase was retained in the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnet even after sintering using the SPS method.

In the Nd-Fe-B magnets, the crystallographic alignment of the tetragonal Nd<sub>2</sub>Fe<sub>14</sub>B phase in the Nd-Fe-B magnets has been evaluated by the ratio, I(006)/I(0410), of the intensity of the c-axis-related peak (006) to that of the strongest peak (410) [26,27]. As the same token, the crystallographic alignment of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase can be evaluated by the ratio I(006)/I(303). The ratio of I(006)/I(303) was 0.27 for the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnet measured perpendicular to the direction of magnetic alignment, while the ratio of I(006)/I(303) was 10.8 for the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnet measured parallel to the direction of magnetic alignment. This indicates the crystallographic alignment of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> phase in the magnet.

As the coercivity of the magnets was improved by adding a small amount of Zn [20,21], zinc powder and 1% zinc stearate powder were added to the  $Sm_2Fe_{17}N_3$  powder before magnetic alignment. Consequently, the effects of this addition on the density and magnetic properties of the magnets were examined. Figure 14 shows the effects of zinc powder addition on the relative density of the  $Sm_2Fe_{17}N_3$  bulk magnets with 1% zinc stearate powder. Unlike in the case of the zinc stearate powder addition, the relative density of the  $Sm_2Fe_{17}N_3$  bulk magnets decreased from 95.4% to 91.5% as the zinc content increased. This indicates that the addition of zinc powder deteriorated the density of the  $Sm_2Fe_{17}N_3$  bulk magnets with 1% zinc stearate powder.



**Figure 13.** X-ray diffraction patterns of the  $Sm_2Fe_{17}N_3$  bulk magnet with 1% zinc stearate powder measured (a) perpendicular to the magnetic alignment and (b) parallel to the direction of magnetic alignment. The black arrows in the  $Sm_2Fe_{17}N_3$  bulk magnet indicate the magnetic alignment of the  $Sm_2Fe_{17}N_3$  powder.



**Figure 14.** Dependence of the relative density of the  $Sm_2Fe_{17}N_3$  bulk magnets with 1% zinc stearate powder on the zinc content.

Figure 15 shows the effects of zinc powder addition on the remanence and coercivity of the  $Sm_2Fe_{17}N_3$  bulk magnets. Since the 1 wt% zinc stearate powder addition effectively increased the magnetic alignment of the  $Sm_2Fe_{17}N_3$  powder, the  $Sm_2Fe_{17}N_3$  bulk magnets with 1 wt% zinc stearate powder were used as the base magnet. While the remanence slightly decreased as the amount of zinc powder increased, the coercivity considerably increased with increasing amounts. This verifies that the addition of zinc powder before the magnetic alignment improved the coercivity of the magnets.

Figure 16 shows the hysteresis loop of the magnet with 1 wt% zinc stearate and 5 wt% zinc powder. The hysteresis loop of the magnet without additives is also shown. The magnet with 1 wt% zinc stearate and 5 wt% zinc powder exhibited a wider hysteresis loop than the magnet without additives, indicating that the use of small amounts of zinc stearate powder and zinc powder increased the magnetic properties of the  $Sm_2Fe_{17}N_3$  bulk magnet. The magnet with 1 wt% zinc stearate and 5 wt% zinc powder showed a high remanence of 86.8 emu/g and a large coercivity of 9.82 kOe.



**Figure 15.** Dependence of the remanence and coercivity of the  $Sm_2Fe_{17}N_3$  bulk magnets on the zinc content.  $Sm_2Fe_{17}N_3$  bulk magnets with 1 wt% zinc stearate powder were used as the base magnet.



**Figure 16.** Hysteresis loops of the  $Sm_2Fe_{17}N_3$  bulk magnets: (a) without additives, and (b) with 1 wt% zinc stearate and 5 wt% zinc powder. The magnets were prepared at 673 K under an applied pressure of 100 MPa for 300 s using the SPS method.

Compared with the hysteresis loop of the  $Sm_2Fe_{17}N_3$  powder (see Figure 6), the remanence and coercivity of the  $Sm_2Fe_{17}N_3$  bulk magnets with 1 wt% zinc stearate powder were slightly smaller than those of the  $Sm_2Fe_{17}N_3$  powder. Thus, further work is still necessary to improve the magnetic properties of the  $Sm_2Fe_{17}N_3$  bulk magnets.

### 3. Materials and Methods

 $Sm_2Fe_{17}$  alloy powder and  $Sm_2Fe_{17}N_3$  powder, prepared by the nitrogenation of  $Sm_2Fe_{17}$  alloy powder, were supplied by Sumitomo Metal Mining Co., Ltd. (Tokyo, Japan). The powder was poured into a BN-coated carbon die and magnetically aligned using an electromagnet before being subjected to spark plasma sintering.  $Sm_2Fe_{17}N_3$  bulk magnets were produced from the  $Sm_2Fe_{17}N_3$  powder using the SPS method under the applied pressure of 100 MPa using a spark plasma sintering apparatus (Plasman, S. S. Alloy, Hiroshima, Japan). The SPS method made the sintering in a vacuum at temperatures between 473 K and 673 K for 300 s. The specimens were cooled in the SPS chamber in a vacuum to room temperature. In order to improve the magnetic properties of the magnets,

zinc stearate powder (0–3 wt%) and zinc powder (0–5 wt%) were added to the  $Sm_2Fe_{17}N_3$  powder, and the mixed powders were magnetically aligned before sintering using the SPS method.

The properties of the original powders and the magnets produced using the SPS method were examined. The morphology of the original powders was investigated using a scanning electron microscope (JSM-IT300LA, JEOL, Tokyo, Japan). The densities of the magnets were measured using the Archimedes method using an electronic balance (GR-120, AND, Tokyo, Japan). The specimens were characterized by X-ray diffraction using Cu K $\alpha$  radiation using an X-ray diffractometer (MiniFlex600, Rigaku, Tokyo, Japan) and magnetic measurements by a vibrating sample magnetometer (BHV-525RSCM, Riken Denshi, Tokyo, Japan). Before magnetic hysteresis measurements of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder were performed, the powder was magnetically aligned and mixed with paraffin. To measure the magnetic properties of the Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets, rod-like specimens ( $1 \times 1 \times 10$  mm) were measured parallel to the length direction to avoid demagnetization correction.

## 4. Conclusions

Although Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder was magnetically anisotropic and possessed high remanence and coercivity, Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets produced from Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> powder using the SPS method exhibited deteriorated remanence and coercivity. Herein, Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets were made using the SPS method with the addition of zinc stearate powder and zinc powder. It was found that the magnetic anisotropy of the magnets was enhanced upon the addition of zinc stearate powder, and the coercivity of the magnets was enhanced upon the addition of zinc powder. The Sm<sub>2</sub>Fe<sub>17</sub>N<sub>3</sub> bulk magnets with 1 wt% zinc stearate powder and 5 wt% zinc powder exhibited a remanence of 86.8 emu/g and coercivity of 9.82 kOe.

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