High performance Sm-Fe-N magnets prepared by compression shearing method

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Abstract. Sm-Fe-N anisotropic magnets were produced from magnetically aligned green compact at room temperature in ambient atmosphere by the compression shearing method. X-ray diffraction studies revealed that the magnets retained their original Sm$_2$Fe$_{17}$N$_3$ phase structure without any appreciable decomposition of the phase and that the phase had a pronounced crystallographic alignment. The magnetic properties of the Sm-Fe-N anisotropic magnets were dependent on the magnetic alignment of the Sm-Fe-N powder before implementation of the compression shearing method. A Sm-Fe-N anisotropic magnet with the magnetic alignment produced under an applied field of 1.6 MA/m exhibited a maximum energy product of 161 kJ/m$^3$ with a high coercivity of 0.88 MA/m.

1. Introduction
With the advent of high-performance Nd-Fe-B permanent magnets, research and development of new permanent magnets has been largely concentrated on rare-earth-based alloys [1-3]. As a result of intensive research, several new rare-earth-based alloys for permanent magnets have been found [4-7]. Among them, Sm-Fe-N alloy is the most promising candidate for high-performance permanent magnets [4]. The superiority of Sm-Fe-N magnets arises from Sm$_2$Fe$_{17}$N$_3$ intermetallic compound, which exhibits high saturation magnetization with a large anisotropy field and a high Curie temperature [4]. Sm$_2$Fe$_{17}$N$_3$ intermetallic compound has been prepared by the production of Sm$_2$Fe$_{17}$ alloy powder and subsequent nitrogenation of the powder by a gas-solid reaction. The resultant Sm$_2$Fe$_{17}$N$_3$ intermetallic compound has thus been produced in powder form.

In the production of high-performance Nd-Fe-B permanent magnets, Nd-Fe-B alloy powder has been consolidated into green compact and then sintered at a high temperature [2]. However, this sintering technique cannot be applied to the production of Sm-Fe-N bulk magnets because Sm$_2$Fe$_{17}$N$_3$ intermetallic compound is not stable at high temperatures and decomposes into $\alpha$-Fe and SmN above 500-600°C [4]. Up to now, Sm-Fe-N bulk magnets have been produced by shock compaction such as by explosion or a gun method [8,9]. Sm-Fe-N bulk magnets with porosities of 2-10% have been realized by optimizing the shock compression conditions [9]. There has been no practical application of this technique, however, due to the difficulties involved.

A recently developed consolidation technique, called compression shearing, can consolidate powders into bulk form at room temperature by the application of shear stress to them [10]. This makes the compression shearing method advantageous for the production of bulk materials from Sm-
Fe-N powders compared with the conventional consolidation techniques. We have recently produced Sm-Fe-N bulk magnets by the compression shearing method [11,12]. The reported magnetic properties, however, were not entirely satisfactory. The magnetic properties of Sm-Fe-N magnets produced by the compression shearing method depend on several processing conditions, particularly the magnetic alignment of the Sm-Fe-N powder. In this study, the effect of the applied field on the magnetic alignment of the Sm-Fe-N powder before implementation of the compression shearing method was investigated. The magnetic properties of Sm-Fe-N magnets produced by the compression shearing method are discussed here.

2. Experiment
Commercially available Sm$_2$Fe$_{17}$N$_3$ powder was used in the experiment. The average particle size of the powder was about 3 μm. The powder was placed on a hardened steel plate and then pressed into green compact under an applied magnetic field of up to 1.6 MA/m before implementation of the compression shearing method. Shear stress was applied to the magnetically aligned green compact by the compression shearing method. Details of the compression shearing method are described elsewhere [11]. The phases of the specimens were examined by X-ray diffraction (XRD) using Cu Kα radiation. The magnetic properties of the specimens were measured using a vibrating sample magnetometer (VSM) with a maximum applied field of 2.0 MA/m. The specimens had been magnetized in a pulsed field of 4.0 MA/m prior to the magnetic measurements. The magnetic measurements were made in the direction parallel to the direction of magnetic alignment. No demagnetization correction was made in the curve. In the calculations of magnetization, the value obtained by Archimedes’ method was used for the density of the specimen.

3. Results and discussion
Figure 1 shows a view of the Sm-Fe-N magnet produced from magnetically aligned green compact under an applied magnetic field of 1.6 MA/m at room temperature in ambient atmosphere by the compression shearing method. Virtually the same specimens were obtained from green compact without magnetic alignment and from magnetically aligned green compact produced under applied magnetic fields of 0.4-1.2 MA/m. The Sm-Fe-N powder was successfully consolidated into bulk magnets at room temperature in ambient atmosphere by the compression shearing method regardless of the applied magnetic field. The resultant magnets were rectangular in shape with dimensions of 18 x 10 mm and a thickness of 0.2 mm. No significant voids or cracks were observed in the magnets, which had a smooth metallic surface.

Archimedes’ method was used to confirm the density of the Sm-Fe-N magnets produced by the compression shearing method. Figure 2 shows the dependence of the density of the Sm-Fe-N magnets on the applied magnetic field before implementation of the compression shearing method. The density of the Sm-Fe-N magnets increased as the applied magnetic field increased, from 7.02 Mg/m$^3$ (91.5% of the X-ray density of the Sm$_2$Fe$_{17}$N$_3$ phase) for the specimen produced without an applied magnetic field to 7.30 Mg/m$^3$ (95.1% of the X-ray density of the same phase) for the specimen produced from magnetically aligned green compact under an applied magnetic field of 1.6 MA/m. It was found that

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**Figure 1.** External appearance of Sm-Fe-N magnet produced from magnetically aligned green compact under an applied magnetic field of 1.6 MA/m at room temperature in ambient atmosphere by the compression shearing method.
the application of a magnetic field before implementation of the compression shearing method is effective in increasing the density of bulk magnets produced by the compression shearing method.

Figure 3 shows the XRD patterns of the Sm-Fe-N magnets produced from magnetically aligned green compact before implementation of the compression shearing method. The specimens were measured parallel to the direction of magnetic alignment. The XRD pattern of the Sm-Fe-N magnet produced without magnetic alignment exhibits the typical powder pattern of the Sm$_2$Fe$_{17}$N$_3$ phase. In the compression shearing method, the shear stress applied to the powder during consolidation not only moves the particles so that the cavities are filled, but also causes friction between the particles while closing the gaps between them. The heat generated by the closing of these gaps may affect the Sm$_2$Fe$_{17}$N$_3$ compound, which is unstable at high temperatures and decomposes into $\alpha$-Fe and SmN above 873 K. However, neither the $\alpha$-Fe phase nor the SmN phase were found in the XRD pattern. This indicates that the Sm-Fe-N magnet was produced by the compression shearing method without decomposition of the Sm$_2$Fe$_{17}$N$_3$ phase. On the other hand, the XRD patterns of the Sm-Fe-N magnets produced from magnetically aligned green compact under applied magnetic fields show a pronounced c-axis alignment of the Sm$_2$Fe$_{17}$N$_3$ phase, which is characterized by the prominent (006) peak. This indicates that the resultant Sm-Fe-N magnets retain the magnetic alignment of the Sm$_2$Fe$_{17}$N$_3$ phase in the compression shearing method and thus exhibit the crystallographic alignment of the Sm$_2$Fe$_{17}$N$_3$ phase. It was found that the degree of crystallographic alignment of the Sm$_2$Fe$_{17}$N$_3$ phase—in this case, the degree of crystallographic alignment of the Sm-Fe-N powder—increased as the applied magnetic field increased.

Figure 2. Dependence of the density of the Sm-Fe-N magnets on the applied magnetic field before implementation of the compression shearing method.

Figure 3. X-ray diffraction patterns of the Sm-Fe-N magnets produced from magnetically aligned green compact before implementation of the compression shearing method. The hysteresis loop of a Sm-Fe-N magnet produced without magnetic alignment is also shown.
Figure 4 shows the dependence of the remanence and coercivity of the Sm-Fe-N magnets on the applied magnetic field before implementation of the compression shearing method. The remanence and coercivity of the isotropic Sm-Fe-N magnet produced without an applied magnetic field are 0.72 T and 0.96 MA/m, respectively. Although the coercivity of the Sm-Fe-N magnets slightly decreased as the applied magnetic field increased, their remanence increased. The higher remanence of the Sm-Fe-N magnets produced from the magnetically aligned green compact compared with that of the magnet produced without magnetic alignment confirms that the Sm-Fe-N magnets produced from the magnetically aligned green compact are magnetically anisotropic. The crystallographic alignment of the c-axis of the Sm$_2$Fe$_{17}$N$_3$ phase in the Sm-Fe-N magnets produced from the magnetically aligned green compact gives rise to the observed higher remanence.

Figure 5 shows the hysteresis loop of the Sm-Fe-N magnet produced from magnetically aligned green compact under an applied magnetic field of 1.6 MA/m by the compression shearing method (b). For comparison, the hysteresis loop of a Sm-Fe-N magnet produced without magnetic alignment is also shown (a).

4. Conclusion
Sm-Fe-N powder was successfully consolidated into bulk materials at room temperature by the compression shearing method. The resultant bulk magnets retained the original Sm$_2$Fe$_{17}$N$_3$ phase structure without any appreciable decomposition of the Sm$_2$Fe$_{17}$N$_3$ phase. The degree of crystallographic alignment of the Sm$_2$Fe$_{17}$N$_3$ phase and the remanence of the Sm-Fe-N magnets
increased as the magnetic field applied before implementation of the compression shearing method increased. An anisotropic Sm-Fe-N bulk magnet with a maximum energy product of 161 kJ/m³ was produced from magnetically aligned green compact under an applied magnetic field of 1.6 MA/m by the compression shearing method.

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