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Production of Sm–Fe–N bulk magnets by the spark plasma sintering method with dynamic compression

Tetsuji Saito [a,*], Kotaro Kikuchi [b]

[a] Department of Mechanical Science and Engineering, Chiba Institute of Technology, 2-17-1 Tsudanuma, Narashino, Chiba, 275-0016, Japan
[b] S. S. Alloy Co., Ltd., 3-13-26 Kagamiyama, Higashi-Hiroshima, Hiroshima, 739-0046, Japan

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ABSTRACT

We report a modified spark plasma sintering method, called spark plasma sintering with dynamic compression (SPS-DC). In the present study, $TbCu_7$-type Sm–Fe–N powder was consolidated into bulk materials of up to 823 K using this method. The Sm–Fe–N powder was successfully consolidated into bulk materials at low temperatures of 573–623 K. It was found that the Sm–Fe–N magnets produced at these low temperatures retained the $TbCu_7$ phase without any appreciable decomposition of that phase. The magnetic properties of the Sm–Fe–N magnet produced at these low temperatures were comparable to those of the Sm–Fe–N powder. The Sm–Fe–N bulk magnet produced at 623 K exhibited a high remanence of 103 A m^2/kg with a high coercivity of 0.73 MA/m.

1. Introduction

Samarium–iron–nitride (Sm–Fe–N) magnets have been attracting renewed interest as high-performance permanent magnets that possess magnetic properties comparable to those of neodymium–iron–boron (Nd–Fe–B) magnets [1–3]. The superiority of Sm–Fe–N magnets arises from $Sm_2Fe_{17}N_3$ intermetallic compound. The saturation magnetization of the $Sm_2Fe_{17}N_3$ phase is slightly lower than that of the $Nd_2Fe_{14}B$ phase, but the magnetocrystalline anisotropy field and Curie temperature are much larger than those of $Nd_2Fe_{14}B$ phase. Sm–Fe–N magnets can therefore be applied to motors for hybrid and electric vehicles without the addition of heavy rare-earth elements such as dysprosium (Dy) as is the case for Nd–Fe–B magnets [4–6]. Unlike in the production of Nd–Fe–B magnets, the sintering technique cannot be applied to the production of Sm–Fe–N bulk magnets because Sm–Fe–N powder is unstable at elevated temperatures and decomposes into the α-Fe and SmN phases [7–9]. The production of Sm–Fe–N bulk magnets has therefore been accomplished by shock compaction, high-pressure compaction, or other nonconventional consolidation techniques such as the compression sharing method. In the shock compaction, the Sm–Fe–N bulk materials with the relative density of 90–98% have been produced at room temperature by the shock compaction using a gun method. The Sm–Fe–N bulk materials with the relative density of 92% have been reported by the high-pressure compaction at 673 K with an applied pressure of 1.5 GPa. However, there has been no practical application of these consolidation techniques due to the difficulties involved [10–14].

The recently developed rapid sintering technique, known as the spark plasma sintering (SPS) method can consolidate powders at relatively lower temperatures and with a short consolidating period [15,16]. These characteristics make the SPS method suitable for the production of bulk materials [17–19]. Some attempts have already been made to produce Sm–Fe–N bulk magnets by the SPS method [20–23]. However, magnetic properties comparable to those of Sm–Fe–N powder have not yet been achieved because of the insufficient density of the magnets. In order to improve the density of these magnets, we have modified the SPS method by applying dynamic compression during sintering. The combination of the SPS and continuous dynamic compression enhances the usefulness of the SPS method by making it possible to produce high-density bulk materials at lower temperatures. The purpose of this study was to investigate the possibility of producing Sm–Fe–N bulk magnets by this modified SPS method, called the spark plasma sintering with dynamic compression (SPS-DC).

* Corresponding author.
E-mail address: tetsuji.saito@it-chiba.ac.jp (T. Saito).

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2. Experiment

$TbCu_7$-type Sm–Fe–N powder ($SmFe_{10}N_{0.5}$) was used in the experiment. Samples of the powders were placed in a WC–FeAl–C alloy die and sintered under a vacuum by SPS. Fig. 1 shows a schematic drawing of the setup for the SPS-DC method. The temperature of the specimen was increased from room temperature to the consolidating temperature of 573–823 K over a period of 300 s and then held at the consolidating temperature for 300 s. The temperature was measured using a thermocouple placed on the surface of the specimen. Fig. 2 shows a temperature–pressure–time diagram of the bulk material produced at 573 K by the SPS-DC method. A pressure of 100 MPa was cyclically applied 150 times during sintering. For comparison, the powder was also consolidated into bulk material by the conventional SPS method using static pressing.

The bulk magnets had typical dimensions of 10 mm in diameter and 1–2 mm in thickness. The specimens were cut for further processing and property measurements. The density of the specimens was measured by Archimedes' method. The specimens were examined by X-ray diffraction (XRD) using Cu K_α radiation. The thermomagnetic properties of the specimens were determined by heating them at a rate of 0.16 K/s in a vacuum using a vibrating sample magnetometer (VSM) with an applied field of 40 kA/m. For the magnetic property measurements, specimens of $1 \times 1 \times 10$ mm were prepared and measured parallel to the length direction to avoid the need for demagnetization correction. The magnetic properties of the specimens were measured by VSM with a maximum applied field of 2.0 MA/m. The specimens were magnetized in a pulsed field of 4.0 MA/m prior to the VSM measurements.

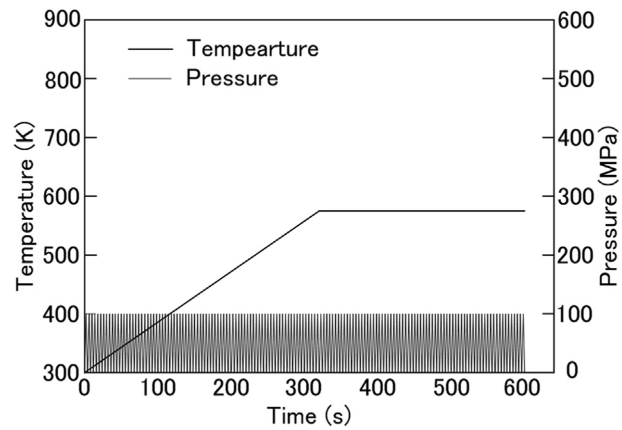

Fig. 2. Temperature–pressure–time diagram of the bulk material produced at 573 K by the SPS-DC method. A pressure of 100 MPa was cyclically applied 150 times during sintering.

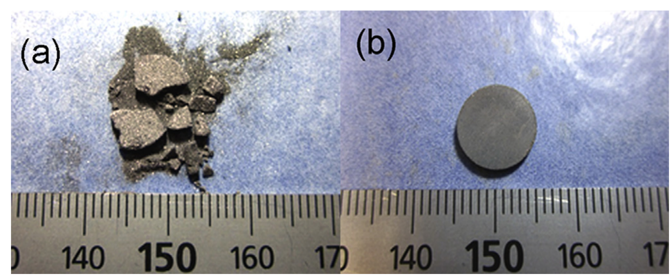

Fig. 3. External appearances of bulk material produced at 573 K by (a) the conventional SPS method and (b) the SPS-DC method.

3. Results and discussion

Sm–Fe–N bulk materials have been produced at temperatures of 673 K or higher by the SPS method [20–23]. In the SPS-DC method described in this paper, Sm–Fe–N bulk materials can be obtained at lower temperatures. Fig. 3 shows the external appearances of bulk materials produced at 573 K by the SPS method and the SPS-DC method. Although the Sm–Fe–N powder was not consolidated into bulk material at such a low temperature by the conventional SPS method, it was successfully consolidated into bulk material at that temperature 573 K by the SPS-DC method. This indicates that the cyclically applied pressure of 100 MPa, in this case 150 times during sintering, is very effective in improving the outcome of the sintering process.

The density of the bulk materials produced by the SPS-DC method is shown in Fig. 4. The density of the bulk material

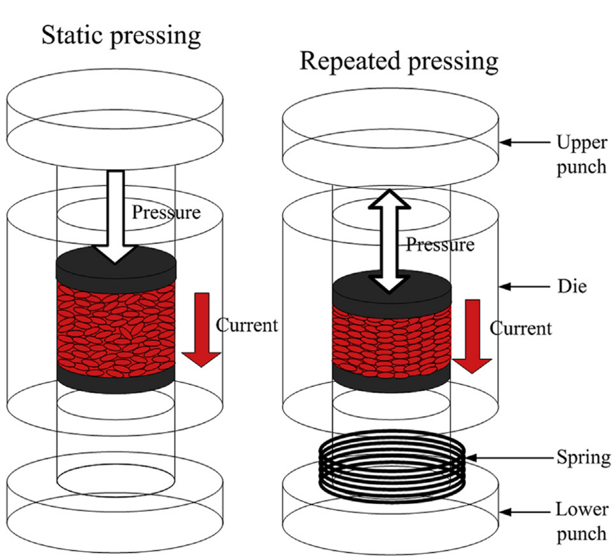

Fig. 1. Schematic diagram showing the setup for the conventional SPS method (left) and the SPS-DC method (right).

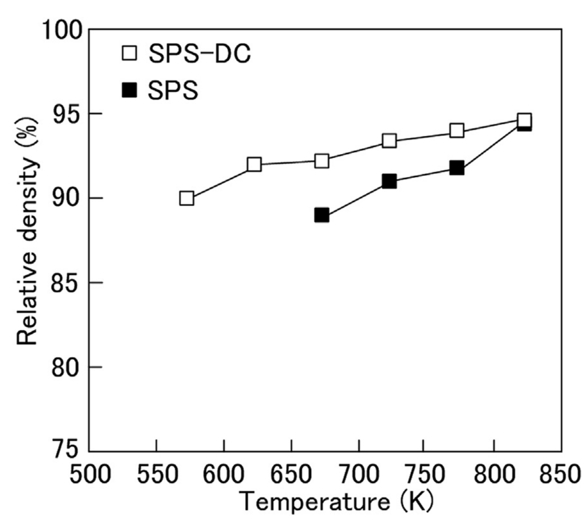

Fig. 4. Dependence of density of bulk materials produced by the SPS-DC method on the sintering temperature. For comparison, the density of bulk materials produced by the conventional SPS method is also shown.