100-μm-thick Nd–Fe–B magnets for MEMS applications produced via a low-temperature sintering route

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Abstract

Magnetic micro-actuators and systems (MAGMAS) require tiny permanent magnets with dimensions of hundreds of micrometers. Such magnets need to have the highest possible energy density, which means Nd–Fe–B magnets are the most appropriate type. Most bottom-up fabrication techniques are either too slow or too expensive; top-down techniques involving machining tend to result in surface damage and a loss of magnetic properties. In this study, we have looked at very neodymium-rich Nd–Fe–B powders that allow us to sinter 100-μm-thick samples to full density at temperatures as low as 800°C. These very thin magnets have coercivities of up to 1000 kA m\(^{-1}\) and are suitable for MAGMAS-type applications.

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1. Introduction

Devices based on micro-electro-mechanical systems (MEMS) are increasingly common; however, most MEMS are micro-sensors, used for detecting changes in, for example, pressure, temperature, and acceleration. Micro-actuators, in contrast, have made relatively slow progress: there are only a few examples of micro-valves, micro-pumps, micro-positioners, etc., because the energy densities of these devices are often too low for the requirements of the application. Magnetic micro-actuators and systems, usually referred to as MAGMAS, are miniature devices that convert electrical energy into mechanical energy, making use of the fields produced by permanent magnets. Such devices could overcome the energy-density problem, provided magnets with thicknesses of 100–500 μm [1] and with properties comparable to bulk magnets can be produced economically.

Thin rare-earth transition-metal (RE-TM) magnets can be produced by familiar techniques like pulsed-laser deposition (PLD) [2], sputtering [3], and molecular-beam epitaxy (MBE) [4]. All of these techniques are able to produce magnets with excellent properties; however, PLD and MBE are probably too expensive for any commercial realization in the area of Nd–Fe–B permanent magnets, but sputtering, plasma spraying [5] (which can deposit material at a practical rate of several μm per minute) and ion–plasma sputtering can produce sufficiently thick magnets with good magnetic properties in several hours or less. An excellent review of some routes for preparing thick-film magnets can be found in Dempsey et al. [6].

The most obvious method for producing small, 100–500-μm-thick magnets is to use a straightforward top-down approach and simply cut up large magnets. Unfortunately, though, high-quality RE-TM magnets can suffer surface degradation during micro-machining, resulting in a loss of coercivity and remanence [6], and Hinz et al. [7] report that the brittleness of Nd–Fe–B magnets “restricts the lower limit of thickness by machining the bulk specimens to some

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hundreds of micrometers”, which eliminates the possibility of producing 100-μm-thick samples using such a process. The same authors, in their own experiments, produced Nd–Fe–B samples from Magnequench’s MQU-F powder with thicknesses in the range 300–600 μm using their one-stage “extreme” die-upsetting process. In comparison, their thinner samples, down to 150 μm, produced using a double die-upsetting process, had diminished coercivities.

Other techniques that have been applied with varying degrees of success include screen printing [8], bonding, and injection molding.

The ideal technique for a MAGMAS magnet would combine a simple, low-cost process with good magnetic properties. For this reason, we have looked at attempting to sinter materials with sufficient amounts of rare earth to overcome the problems of evaporation that are always associated with sintering small samples at temperatures above 1000 °C. Whereas, previously reported sintering-type techniques have involved complex processing techniques like two-stage die upsetting while applying direct Joule heating to produce 200-μm-thick samples [9] or a special granulation process [10], we have chosen to apply a simple process and modify the starting material to produce high-coercivity RE-TM magnets with thicknesses down to 100 μm.

2. Experimental

The five alloys used in this series of experiments were produced by induction melting appropriate amounts of neodymium (99.9%), iron (99.9%) and standard Fe2B ferro-boron, and pouring each molten alloy into a copper book-type mold with dimensions of 40 mm × 250 mm × 400 mm. Each 15-kg melt was poured at 1530 °C. Small samples from close to the edge of each ingot were mounted and prepared for metallographic examination. Approximately 5 kg of each ingot was then hydrogen decrepitated and prepared for metallographic examination. Approximately 5 kg of each ingot was then hydrogen decrepitated and then milled in argon to a mean particle size of 5 μm. The chemical compositions of the five alloys are shown in Table 1.

The powders were stored under argon prior to pressing in parallel and perpendicular presses: the latter with a transverse magnetic field of 0.75 T. The amount of powder poured into the pressing die was varied so as to produce samples with thicknesses in the range 100 μm–5 mm. The pressure applied to form the green compacts was 390 MPa.

The green compacts were heated in a vacuum of approximately 10⁻³ mbar at 5 °C min⁻¹ to temperatures in the range 680–960 °C for 1 h, after which they were allowed to cool in the furnace to room temperature. The initial cooling rate was limited to 10 °C min⁻¹. Each sintering experiment involved 3–5 pressed green samples that were placed in a tantalum sample tube, to prevent contact with the silica-glass furnace tube, and located precisely in the hot zone of the furnace.

After the sintering treatment in the furnace the samples were cut with a diamond-wire cutter so as to have sides of approximately 250 μm × 250 μm and then measured at room temperature using a vibrating-sample magnetometer (VSM) with a maximum field of 1.8 T to provide information on their magnetic properties.

The chemical analyses of the phases in the as-cast alloys were carried out with a JEOL 840A scanning electron microscope.

3. Results and discussion

The microstructures of the five cast alloys are shown in Fig. 1. The various phases present in the alloys were analyzed using energy-dispersive X-ray diffraction and wavelength-dispersive X-ray diffraction. The phases are identified in the figure. It is clear that with increasing neodymium concentration in the as-cast alloys there is a pronounced increase in the amount of Nd-rich material, and due to the richness of the starting compositions in terms of neodymium all the alloys were in the two-phase region of the phase diagram, where φ (Nd2Fe14B) is in equilibrium with the liquid; this is in contrast to conventional Nd–Fe–B magnet compositions, where a three-phase region exists and φ is in equilibrium with the liquid and η (Nd1.1Fe5B3) [11]. The proportion of coarse particles of the primary φ tetragonal phase is reduced with the extra neodymium, so that there is substantially more Nd-rich matrix already in alloy 2. Within this matrix there is also an increased share of binary eutectic (Nd + φ). These same features are also clearly present in alloy 3, with 30.9% Nd. The higher neodymium concentration in the alloys 4 and 5 and the non-equilibrium solidification result in increasingly smaller amounts of φ phase and separated solidification of the Nd-rich residual melt. The consequence of this is the presence of primary crystals of neodymium and a binary eutectic (Nd + φ).

Fig. 2 shows a graph of the effect of the sintering temperature on the intrinsic coercivity of the samples produced in the range 680–960 °C. Each sample was produced from 0.8 g of material and pressed in a die with a diameter of 6 mm. The thicknesses of the samples that fully densified were approximately 5 mm. It is clear that the samples produced from alloys 1 and 2 had very poor properties, and that these compositions, which are relatively close to the classical Neomax composition of Nd1.3Fe77B20, require much higher temperatures to densify and develop a high coercivity. In contrast, the samples

Table 1

<table>
<thead>
<tr>
<th>No.</th>
<th>Chemical composition</th>
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<tbody>
<tr>
<td>1</td>
<td>Nd18.3Fe72.8B9.8</td>
</tr>
<tr>
<td>2</td>
<td>Nd31.1Fe68.9B0.4</td>
</tr>
<tr>
<td>3</td>
<td>Nd30.6Fe64.1B5.3</td>
</tr>
<tr>
<td>4</td>
<td>Nd31.2Fe58.8B1.0</td>
</tr>
<tr>
<td>5</td>
<td>Nd51.0Fe40.3B8.7</td>
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from alloys 3–5 exhibited excellent coercivities of 800–1000 kA m\(^{-1}\) in the range 800–960 °C. The plateau in the graph between 800 and 960 °C is, however, a little deceptive: although the measured magnetic properties of the samples remained high, in terms of coercivity, the physical characteristics deteriorated rapidly with the samples beginning to deform and melt at temperatures above 840 °C (alloy 5), 880 °C (alloy 4) and 960 °C (alloy 3).

At lower temperatures, i.e., below 750–800 °C, the coercivity of the samples from alloys 3 and 4 fell off rapidly as the materials failed to densify completely. Samples from alloy 5, however, retained a coercivity of 800 kA m\(^{-1}\), at least down to 680 °C. The fall off in coercivity is not a direct consequence of the lack of density. However, the very high ratio of the surface area to the volume means that evaporation rates for the neodymium are high, much higher than with a conventional bulk magnet, and this tends to lead to a shortage of rare-earth-rich phase, which is required to encompass the individual grains and fully densify the sample. The consequence of this can be a porous structure with a less-than-perfect coating of the grains and a tendency to form small oxides and defects on the surfaces of the Nd\(_2\)Fe\(_{14}\)B grains, thus leading to a lower coercivity.

Fig. 1. Optical micrographs of the as-cast microstructures of samples 1–5 (\(\phi = \text{Nd}_2\text{Fe}_{14}\text{B}\)).

Fig. 2. Effect of processing temperature on the intrinsic coercivity.

Fig. 3. Effect of processing temperature on the remanent magnetization.
The results of the measurements of the same samples' remanent magnetization after magnetizing in a field of 1.8 T are shown in Fig. 3. The curves are similar to the coercivity curves in Fig. 2; this is not surprising as the same inability to densify at low temperatures that reduced the coercivity of the samples from alloys 3 and 4 also results in a decrease in the remanent magnetization of the samples.

Fig. 4 shows the microstructures from samples of alloys 3 (Fig. 4a) and 4 (Fig. 4b) that were sintered at 800 °C. There is clear evidence of porosity and a lack of full density in Fig. 4a.

On the basis of the results shown in Figs. 2–4 we chose a sintering temperature of 800 °C and produced samples from alloys 3–5 with thicknesses in the range 100 μm–5 mm. This thickness variation was achieved by altering the amount of powder added to the die between 20 mg and 0.8 g. The results of the measurements of the intrinsic coercivity for different sample thicknesses are shown in Fig. 5. It is clear that all three alloys can produce magnets with good properties when the samples are relatively thick, i.e., 1 mm or more; however, as the samples are made thinner, below 0.5 mm, the coercivities of the samples from alloy 3 decline rapidly, while those of alloy 4 remain constant, and those from alloy 5 were observed to increase slightly.

Having determined that we required a composition in the range Nd_{41.0}Fe_{50.4}B_{8.6}–Nd_{51.0}Fe_{40.7}B_{8.3} in order to obtain high coercivities for 100-μm-thick samples, we used a perpendicular-alignment press and a field of 0.75 T to produce an aligned magnet from alloy 4 powder. The sample was pressed as the magnetic field was applied and then sintered in vacuum, like the other samples, at 800 °C. The magnetic properties are shown in Fig. 6. A polished cross section of a 100-μm-thick sample from alloy 4 is shown in Fig. 7. It is worth pointing out at this point that an anisotropic magnet with a remanence of, for example, 0.6 T is much more useful than an isotropic magnet with the same remanence. This is to do with the ease of magnetization, a critical factor in systems like MAGMAS, where in-situ magnetization within the system is nearly always a requirement. The applied field required to near-saturate an isotropic Nd–Fe–B-type magnet is much higher than that for the case of an aligned sample. This can be a problem with systems where the magnets have to be mounted in an unmagnetized state and later magnetized, but particularly so with miniaturized systems.

The microstructure in Fig. 7 demonstrates how effective the process of low temperature sintering a high-Nd-content alloy is for producing extremely thin samples without any evident distortion. The sample shrinks during the sintering by about 15%, but still maintains a relatively smooth surface and a parallel cross section. The higher magnification microstructure in Fig. 4b shows how the rare-earth-rich material matrix evenly surrounds the grains, providing them with good isolation. A close inspection of the \( \phi \) grains reveals the presence of small cracks, a consequence of the hydrogen-decrepitation process. Under normal sintering conditions, i.e., at about 1080 °C, these cracks would disappear as a result of the enhanced diffusion and grain growth; however, at lower temperatures they remain, and

![Fig. 4. SEM microstructure of (a) alloy 3, sintered at 800 °C, and (b) alloy 4, sintered at 800 °C.](image-url)
do not appear to have a significantly detrimental effect on the properties, except, perhaps, some loss of squareness. This method for producing very thin magnets for MEMS applications clearly has potential. First of all it is cheap; no amount of high remanence values are of any use if the magnets produced are prohibitively expensive, as would be the case with more exotic techniques like PLD or MBE. Second, the process is familiar; there is no requirement for the introduction of new techniques or the purchase of expensive new equipment. Thirdly, the process is not intrinsically isotropic, as is the case with many other techniques.

Further investigations involving non-hydried materials and some modest changes in the chemistry will be undertaken in an attempt to improve the squareness and the remanence of these thin Nd–Fe–B magnets. Our future work in this area will also focus on looking at compositional modifications that will benefit both the sintering process and the magnetic properties. Our aim will be to develop a highly anisotropic sintered magnet with a thickness of just 50 μm.

4. Conclusions

It is clear from this investigation that the increase in the amount of neodymium-rich constituents directly improves the magnetic properties of 100-μm-thick magnets. By applying a low temperature sintering process and using very neodymium-rich Nd–Fe–B starting alloys we have been able to produce extremely thin, relatively undeformed permanent magnet samples of 100-μm thickness using a simple press-and-sinter process. These magnets are fully dense and exhibit excellent coercivities up to 1000 kA m⁻¹ without the addition of anisotropy-enhancing elements like dysprosium. When cut to shape these magnets are suitable for magnetic micro-actuators and systems (MAGMAS).

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References