Nd–Fe–B permanent magnets fabricated by low temperature sintering process

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ARTICLE INFO

ABSTRACT

Fabrication of 13Nd–2Dy–6B–bal. Fe (at.%) sintered magnets by low temperature sintering was demonstrated, focusing on grain size control. A magnet with a density higher than 99% was successfully obtained by sintering a Nd–Fe–B powder compact at 970 °C for 20 h. The average grain size of the magnet was 5.5 μm, which was similar to that of the initial powder size (5 μm). Compared with a conventionally sintered specimen (1070 °C/4 h), the coercivity of the magnet increased from 1672 kA/m to 1823 kA/m upon low temperature sintering. Consequently, the low temperature sintering process enabled us to suppress grain growth and to distribute the grains more uniformly, which resulted in improved magnetic properties, in particular improved intrinsic coercivity. The grain-size reduction through the low temperature sintering was explained by analyzing microstructures in relation to sintering behavior.

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1. Introduction

Sintered magnets are core materials of variable advanced industries from electronic vehicles to medical devices because of their outstanding magnetic properties [1–3]. However, typical sintered Nd–Fe–B magnets are limited in application due to low Curie temperature and thermal coercivity degradation at elevated temperatures. For the utilization of sintered Nd–Fe–B magnets at elevated temperatures, thermal degradation must be overcome by higher coercivity. The coercivity of Nd–Fe–B sintered magnets is determined through reversal domain nucleation in applied magnetic fields. The intrinsic coercivity can be expressed as [4]

$$_iH_c = \alpha H_A - N_{eff}M_s$$

where H_A is an anisotropy field, equal to $2K_1/M_sN_{eff}$ is a demagnetizing parameter, and α is the degradation constant of the magnetic anisotropy caused by defects in the grain boundaries or surface [5]. The constants α and N_{eff} are determined mainly by the sizes and shapes of grains, the grain boundary, and defects in the areas of domain nucleation. Grain refinement is very effective for increasing coercivity due to the inhibition of domain wall motion in applied fields [6]. Nd–Fe–B sintered magnets are composed of the $Nd_2Fe_{14}B$ hard magnet phase as a matrix and the Nd-rich phase at the grain boundary, and are manufactured by liquid-phase sintering of the Nd-rich liquid phase area [7,8]. Liquid-phase sintering is a

densification method of powder compacts containing more than one component at a temperature above the solidus line of one of the components. Liquid-phase sintering plays an important role in the production of rare-earth magnets. However, grain growth and abnormal grain growth are present during liquid-phase sintering [9,10].

In this study, we demonstrated a low temperature sintering (LTS) process to achieve higher coercivity via grain refinement. The increase in coercivity of the Nd–Fe–B sintered magnet is discussed from a microstructural perspective.

2. Experimental procedures

A starting alloy with a composition of 13Nd–2Dy–6B–bal. Fe (at.%) was prepared by strip casting after materials were melted in a vacuum atmosphere. Nd–Fe–B alloy strips were jet-milled in a N_2 atmosphere to produce a powder with an average particle size of 5.0 μm. The powder particles were magnetically aligned at 2.0 MA/m and compacted at 50 MPa to dimensions of $15 \times 10 \times 12$ mm³. The green compact was sintered non-isothermally while increasing sintering temperatures from 700 °C to 1100 °C under a vacuum of $<1.2 \times 10^{-5}$ torr, and then the sintering behavior was observed. For the LTS process, the compacts were heated to 950–970 °C, respectively, at a heating rate of 10 °C/min. The compacts were then held at each temperature for as long as 20 h. For comparison, conventional sintering was carried out at the same heating rate, at 1070 °C for 4 h under a vacuum of $<1.2 \times 10^{-5}$. Initial particle sizes were analyzed using a particle size analyzer (PSA). The densities of sintered samples were measured according to the Archimedes principle. The grain sizes of the sintered samples were evaluated using an image analyzer (UTHSCSA Image Tool) after polishing a cross section of the samples. The fractographs of the samples were observed with an optical microscope (OM) and a scanning electron microscope (SEM). Phases in sintered specimens were analyzed with an X-ray diffractor (XRD). The magnetic properties of the specimens were measured with a B-H loop tracer (Magnet-Physik Permagraph C-300) at room temperature.

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3. Results and discussion

The average particle size of the initial (Nd,Dy)–Fe–B powder was 5 μm with a distribution of 1–10 μm. To observe sintering behavior, green compacts of this powder with a relative density of around 58% were prepared through uniaxial pressing at 50 MPa. The compacts were then heated to 700–1100 °C at a heating rate of 10 °C/min and sintered non-isothermally. The relative densities and average grain sizes of the sintered specimens are shown in Fig. 1(a). The relative density began to increase at above 900 °C, while the grain size began to increase above 1000 °C with an abrupt increase above 1070 °C, reaching a value of around 7.3 μm at 1100 °C.

Densification was accelerated above 950 °C because the liquid volume in the specimen was increased and the liquid flows were relatively high. Rapid grain growth above 1070 °C may have been due to excessive thermal energy left over from the densification process. When closed pores were formed inside a sintered body, the densification process slowed and most of the thermal energy was consumed by grain growth. In order to reduce such grain growth as much as possible, we attempted to sinter the specimens in the temperature range of 950–1000 °C, at which liquid flows would be sufficient. In liquid phase sintering, the use of the proper holding time at the specified temperatures in liquid state accelerates the densification process [9].

Fig. 1(b) shows the relative density of the specimens sintered at 950 °C and 970 °C for up to 20 h. For comparison, the relative densities of the specimens sintered at 1070 °C are also shown. When the specimens were held for 20 h at 950 °C, the relative density reached 94% and relative densities approaching full density were obtained at temperatures higher than 970 °C, depending on holding time. When the sintering temperature was lower than 950 °C, the liquid phase volume was too small to spread through the structure. Above 970 °C, a fully dense sintered body could be obtained if the holding time was sufficient, because the liquid phase volume was large. Based on the results shown in Fig. 1(a) and (b), the optimum sintering temperature was 970 °C, not exceeding 1000 °C.
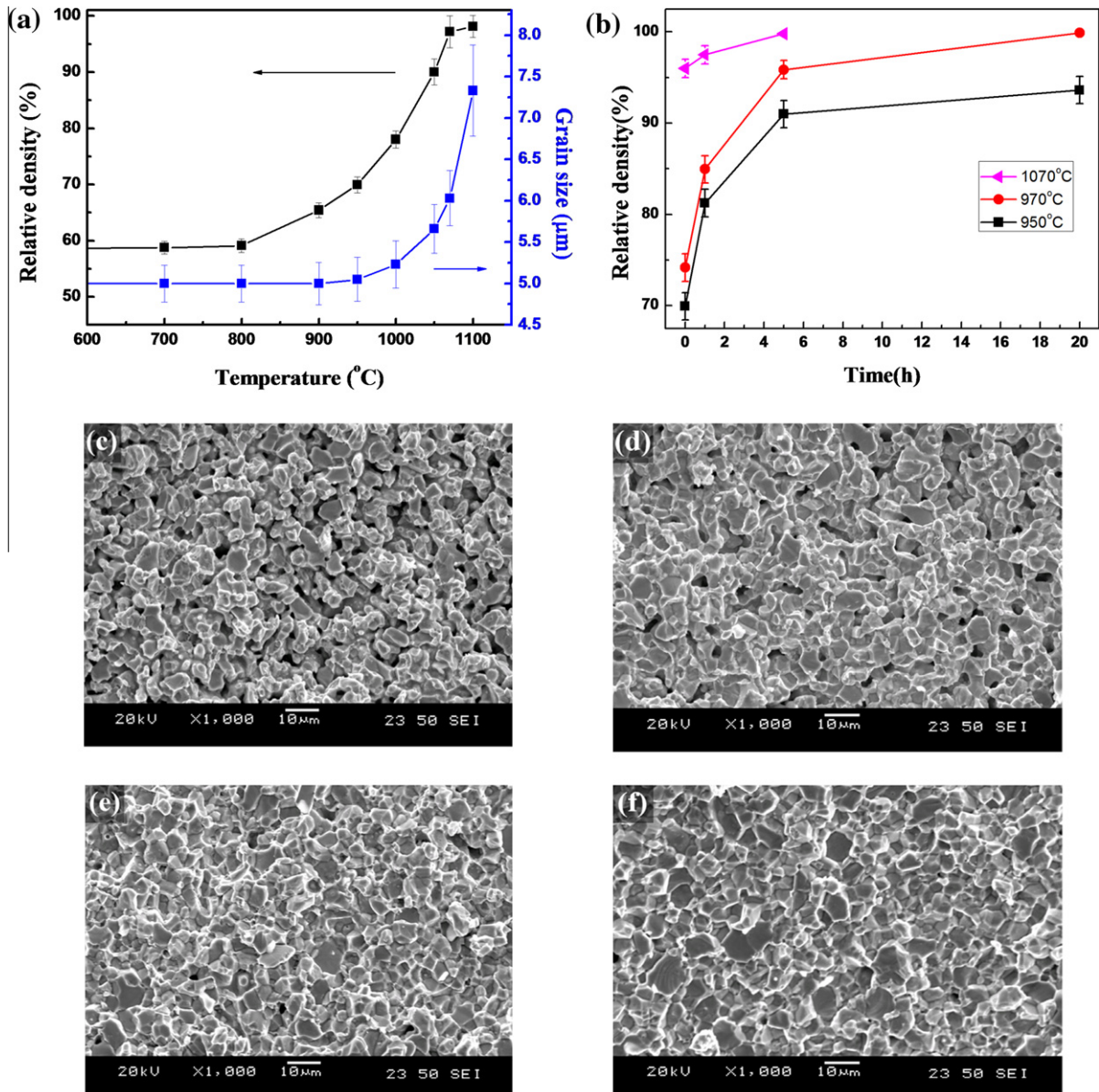

Fig. 1. (a) Changes in relative density and grain size of non-isothermally sintered (Nd,Dy)–Fe–B. (b) Changes in relative density of sintered (Nd,Dy)–Fe–B as a function of holding time. Fractographs of (Nd,Dy)–Fe–B isothermally sintered at 970 °C for (c) 0 h, (d) 1 h, (e) 5 h, and (f) 20 h.