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Anisotropic HDDR-treated Nd–Fe–B alloy flakes for mechanically oriented composite magnets

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Abstract

To explore the possibilities of producing Nd–Fe–B materials for the application of mechanically oriented composite magnets, $Nd_{12.6}Fe_{62.3}Co_{17.4}Al_{0.6}Zr_{0.1}B_{7.0}$ alloy flakes were prepared by hydrogenation–decomposition–desorption–recombination (HDDR)-treating the melt-spun ribbons, and magnetic properties of the resin-bonded compacts made from them were examined. The as-spun ribbons of 230 μm in thickness exhibit a texture with the *c*-axes of $Nd_2Fe_{14}B$ grains oriented perpendicularly to the ribbon planes, and possess low coercivities (H_{cJ}'s) of 90 kA/m. HDDR treatment can induce high H_{cJ}'s of approximately 880 kA/m in these ribbons maintaining the original crystallographic orientation. The flakes obtained by crushing the HDDR-treated ribbons can be mechanically aligned by compaction molding with no applied magnetic field. The resin-bonded compact prepared from the flakes with the highest magnetic anisotropy exhibits B_r of 0.78 T and $(BH)_{max}$ of 106 kJ/m^3 in the pressing direction (easy magnetization direction), when its density is 6.0 Mg/m^3. These magnetic properties are superior to those of the isotropic resin-bonded magnets produced from the commercially available rapidly quenched Nd–Fe–B powders.

Keywords: Nd–Fe–B alloy flake; Melt-spinning; HDDR; Mechanical orientation; Anisotropy; Coercivity

1. Introduction

The hard ferrite plate-like particles of which the easy magnetization directions are normal to their major axes, are widely used in anisotropic composite magnets. They are mixed with rubber and formed into sheets by an extrusion or rolling method. Since these particles have high aspect ratios (major axis length/minor axis length), their easy magnetization directions are aligned perpendicularly to the composite sheet planes by shear force in the forming process. For the production of the anisotropic composite magnets, the mechanical orientation method is more cost-effective than the process using an aligning magnetic field because of the high productivity and elimination of the particular equipments for generating the magnetic field.

Although, in Nd–Fe–B system, anisotropic resin-bonded magnets produced by field orientation of the HDDR-processed powders are now commercially available [1,2], no reports have been published on fabricating mechanically oriented bonded magnets so far. Several attempts have been made to produce magnetically anisotropic Nd–Fe–B alloy flakes, which would be the promising materials for mechanically oriented magnets, by using the melt spinning method [3–8], and remanence (B_r) of around 0.9 T and energy product ($(BH)_{max}$) of up to 88 kJ/m^3 have been reported for the as-spun flakes [4,7]. These values are much smaller than the theoretical values of magnetic $Nd_2Fe_{14}B$ phase (1.6 T and 509 kJ/m^3, respectively). The main reason is that the relatively small quenching rates which are favorable for developing the correlated *c*-axis (easy magnetization axis) orientation

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of $Nd_2Fe_{14}B$ phase in the flakes, produce coarse $Nd_2Fe_{14}B$ crystallites resulting in small coercivities (H_{cJ}'s) of typically less than 480 kA/m [4,7]. In this study, to explore the possibilities of producing anisotropic Nd–Fe–B magnet materials suitable for mechanical orientation, preparation conditions of the melt-spun flakes with strong crystallographic texture were investigated in detail, and then they were subjected to a hydrogenation–decomposition–desorption–recombination (HDDR) treatment. The HDDR treatment can produce fine recombined $Nd_2Fe_{14}B$ grains of approximately 0.3 μm in diameter from the original large grains maintaining the initial crystallographic orientation [1]. It is expected that this treatment can raise H_{cJ}'s in the textured flakes, and consequently improve their B_r's and $(BH)_{max}$'s.

2. Experimental procedure

The mother alloy buttons with the composition of $Nd_{12.6}Fe_{62.3}Co_{17.4}Al_{0.6}Zr_{0.1}B_{7.0}$ were prepared by plasma-arc melting the constituent elements. It was reported that the addition of particular elements such as zirconium [7] and aluminum [8] promotes the preferential growth of $Nd_2Fe_{14}B$ grains with *c*-axes normal to the melt-spun ribbon planes. Cobalt and zirconium are also known as the additive elements that produce highly anisotropic Nd–Fe–B HDDR powders through transmitting the crystallographic orientation of the original $Nd_2Fe_{14}B$ grains to the final recombined grains [1]. Several alloy pieces were introduced into a 10 mm diameter quartz tube with a small diameter orifice of 0.5–0.7 mm at the bottom. They were melted by a high frequency induction furnace and the melts were ejected onto a rotating copper wheel in an argon atmosphere to prepare the alloy ribbons. The surface velocities of the wheel were varied from 2 to 30 m/s. The pressure for ejecting the molten alloys was 20 kPa. The size of the ribbons so obtained was 40–340 μm in thickness and 1–3 mm in width. Some parts of the ribbons were subjected to the HDDR treatment schematically shown in Fig. 1. To obtain alloy flakes with high aspect ratios (length/thickness), hydrogen decrepitation of the ribbons should be avoided in the treatment. In this study, hydrogen gas was introduced into the furnace at 820 °C where the hydrogenation–decomposition reaction proceeds without cracking. This type of treatment was referred to as solid-HDDR by Gutfleisch et al. [9]. After hydrogenation for 50 min, hydrogen gas in the furnace was replaced by argon gas, and the ribbons were exposed to atmospheric argon gas for 4 min. This intermediate argon treatment followed by evacuation to a high vacuum can enhance the correlated *c*-axes orientation of the recombined grains [10]. The as-spun and HDDR-treated ribbons so produced were crushed into flakes with aspect ratios larger than three. The length of the flakes varied from 0.2 to 2 mm depending on the ribbon thickness. The flakes were mixed with epoxy resin and compacted into cubes with an edge of 12 mm at a pressure of 600 MPa under no applied orienting field. The resin-bonded compacts were hardened at 100 °C for 1 h.

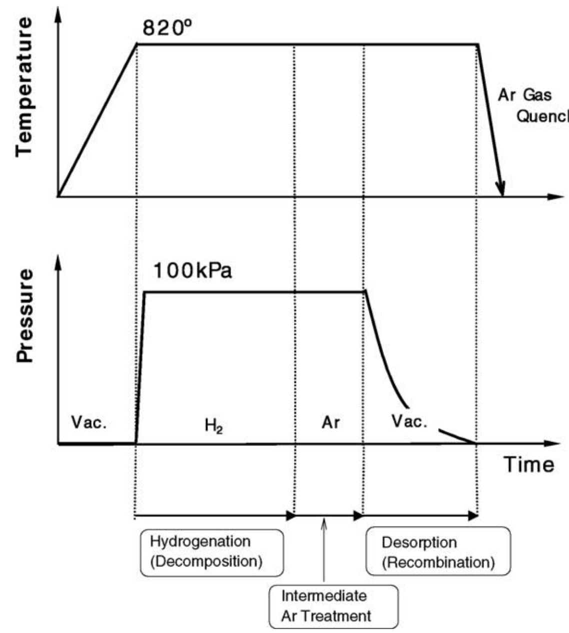

Fig. 1. Schematic diagram of the HDDR treatment.

Densities of the compacts were approximately 6.0 Mg/m^3. Magnetic properties of the compacts were measured with a *B–H* curve tracer after magnetizing them in a pulsed magnetic field of 5570 kA/m. The microstructures of the ribbons were examined in a scanning electron microscope (SEM).

3. Results and discussion

Fig. 2 shows a cross-sectional view of the resin-bonded compact made from the as-spun alloy flakes of 230 μm in thickness. It can be seen that most of the flake planes are aligned perpendicularly to the pressing direction. The occurrence of mechanical orientation of other as-spun and HDDR-treated flakes of this study was also confirmed in their resin-bonded compacts. Hence, the magnetic anisotropy of the

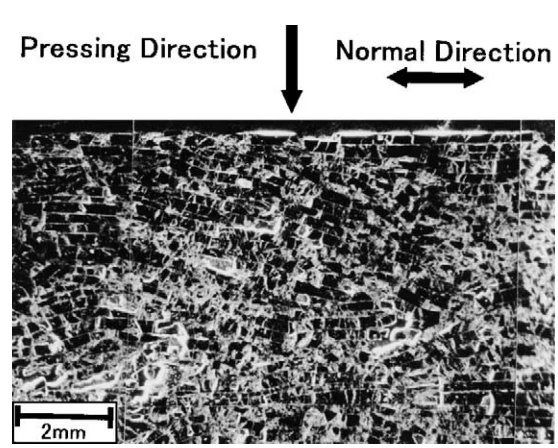

Fig. 2. Cross-sectional view of the resin-bonded compact made from the as-spun Nd–Fe–Co–Al–Zr–B flakes of 230 μm in thickness.