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The effect of key process parameters on crystallographic texture and magnetic properties of PrFeB HD sintered magnets produced using high-energy milling

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Abstract

A combination of hydrogen decrepitation and planetary ball milling has been used to produce sintered $Pr_{16}Fe_{76}B_8$ magnets. The effects of key process parameters and texture have been studied. The alignment degree ($\langle \cos \Theta \rangle$) has been investigated by X-ray pole figure analysis using the (0 0 4) reflection. Magnets prepared from the alloy decrepitated for 3.6 ks and milled for 1.8 ks exhibited a low degree of crystal alignment (0.71 ± 0.02) due to polycrystalline particles. Increasing the milling time to 4.5ks has led to an improvement in $\langle \cos \Theta \rangle$ to 0.84 ± 0.02. This has been ascribed to the smaller particle size with a narrower size distribution as well as a more favorable particle shape for orientation. Superior alignment degree ($\langle \cos \Theta \rangle = 0.88 \pm 0.02$) has been achieved for the sintered magnet from the alloy decrepitated for 120 s and milled for 5.4 ks. This particular processing condition has led to a magnet with $(BH)_{max} = 250 \pm 5\,kJ\,m^{-3}$, value found in magnets produced using roller ball milling, with the advantage of a reduced milling time (about 90%).

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Keywords: Crystallographic texture; High-energy milling; Hydrogen decrepitation; PrFeB; X-ray pole figure

1. Introduction

Crystallographic alignment is essential for achieving high remanence and energy product in sintered magnets. This is accomplished by milling the magnetic alloy and orienting the particles in a strong magnetic field [1]. Magnet texture is dependent on the characteristics of the powder (size, shape, flowability, etc.), which are influenced by the alloy state and the milling conditions. Recently, it has been shown [2] that $Pr_{16}Fe_{76}B_8$ sintered magnets can be produced using the hydrogen decrepitation (HD) process and a planetary ball mill (PBM) with a remarkable reduction in the processing time compared to magnets produced using roller ball milling (RBM) [3]. In the present work, the texture of sintered $Pr_{16}Fe_{76}B_8$ magnets produced using a PBM has been investigated using X-ray pole figure analysis. The hydrogenation and milling steps were used to modify the alignment degree and improve the magnetic properties. In order to establish the optimum texture, a study of the relationship between the alignment degree of the magnets and milling time was carried out for the $Pr_{16}Fe_{76}B_8$ alloy. In an attempt to improve further the anisotropy of the $Pr_2Fe_{14}B$ phase in the HD powder, the hydrogenation time was also modified.

2. Experimental

A commercial $Pr_{16}Fe_{76}B_8$ alloy in the as-cast state was used in this study. Details of the preparation of the sintered magnets and magnetic measurements have all been described in previous papers [2,4]. To produce the standard Pr-based magnets using the HD process, the alloy was exposed to 0.2 MPa of hydrogen for 3.6 ks and milled at 200 rpm. The decrepitated alloy was milled from 1.8 to 5.4 ks. The resultant powder was orientated in a pulsed

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magnetic field of 6 T, isostatically pressed at 200 MPa, vacuum sintered at 1333 K for 3.6 ks and cooled in the furnace ($\sim$10 K min^{-1}). In order to enhance the HD powder anisotropy, the hydrogenation time was also diminished from 3.6 ks to 60, 90 or 120 s. X-ray diffraction (XRD) has been employed to characterize the alloy and HD material using CuKα radiation (2θ was varied between 20° and 70° at a rate of 1° min^{-1}).

The alignment degree of the magnets was determined by X-ray pole figure analysis using the (0 0 4) reflection, following the experimental procedure reported in detail previously [5]. The (0 0 4) normalized intensity data was fitted for a Gaussian distribution and $\langle \cos\Theta \rangle$ was calculated based on the Stoner–Wohlfarth model [6]. Microstructural observations were carried out using a scanning electron microscope (SEM). Grain size measurements of the sintered magnets were carried out using image analysis, and the samples were etched with aqua regia to reveal grain boundaries.

3. Results and discussion

The effects of milling time on the degree of alignment of the standard $Pr_{16}Fe_{76}B_8$ HD magnets are shown in Fig. 1. There is an increase in the degree of alignment of the magnets with milling time and this is due to increase in the proportion of single crystal grains ($Pr_2Fe_{14}B$ phase) obtained by milling. The highest magnetic alignment was achieved in the magnets produced using the HD powder milled for 4.5 ks. Short milling time and/or low milling energy can yield coarse powders with large size distribution (with polycrystalline structure) and domains with different directions of easy magnetization [7] and, hence,

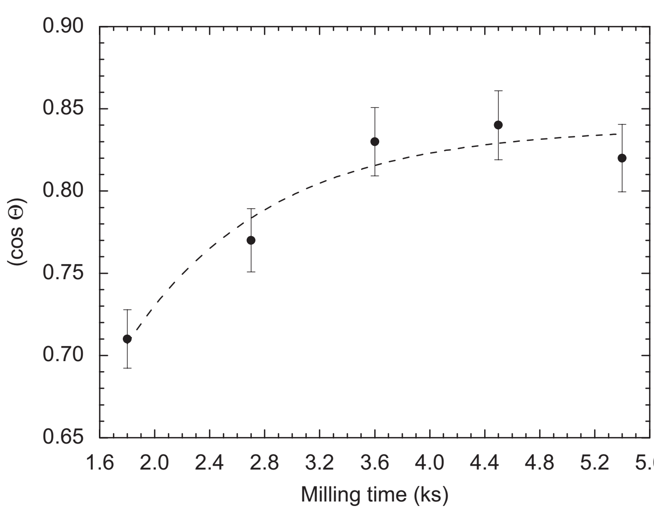

Fig. 1. Variation of the crystallographic texture with milling time for sintered $Pr_{16}Fe_{76}B_8$ magnets produced employing the standard procedure.

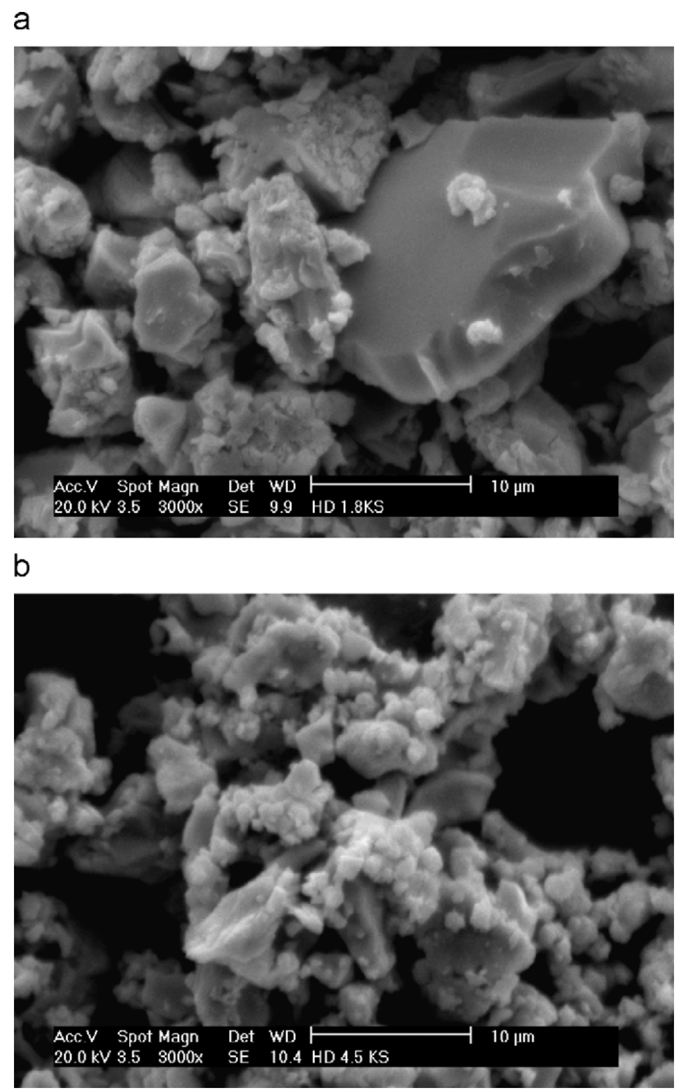

Fig. 2. SEM micrographs of the $Pr_{16}Fe_{76}B_8$ alloy (a) exposed to H_2 for 3.6 ks and milled for 1.8 ks, (b) exposed to H_2 for 3.6 ks and milled for 4.5 ks.

Table 1
Properties of sintered $Pr_{16}Fe_{76}B_8$ magnets produced using high-energy milling [2]

Milling time (ks)	B_r ($\pm$2%) (mT)	$\mu_{0i}H_c$ ($\pm$2%) (mT)	$(BH)_{max}$ ($\pm$2%) (kJ m^{-3})	MGS (µm)	σ (µm)
1.8	860	1190	134	4.96	2.78
2.7	920	1320	162	4.21	2.80
3.6	960	1310	176	3.51	2.00
4.5	1020	1420	200	3.08	1.60
5.4	800	1410	118	2.95	1.68

sintered magnets with low crystal alignment. Longer milling times reduce particle size as well as narrow its size distribution, although overmilling can promote powder agglomeration and reduce alignment. A similar behavior between $\langle \cos \Theta \rangle$ and particle size was reported previously [7]. The magnetic properties, mean grain size (MGS) and standard deviation of the MGS (σ) for the HD magnets are given in Table 1.

The structures of the hydrogenated $Pr_{16}Fe_{76}B_8$ alloy milled for 1.8 and 4.5 ks are shown in Fig. 2(a) and (b), respectively. In general, the former shows an irregular shape as well particles with flat surfaces. It is also possible to identify individual plate-like particles. Due to short milling period, the MGS of this sample is the largest among all conditions evaluated (4.96 μm) and the grain size distribution is the poorest, indicated by the standard deviation. The latter shows particles with a more rounded shape, although irregular particles with flat surfaces are still identified. There was the reduction in the mean particle size, evidenced by the powder agglomeration. Due to longer milling time, the grain size distribution is narrower.

Fig. 3 shows the XRD pattern of the $Pr_{16}Fe_{76}B_8$ alloy in the as-cast state and exposed to hydrogen for 60, 120 s and 3.6 ks. The X-ray pattern obtained for the alloy exposed to hydrogen for 90 s has not been included to avoid overlapping. There is a gradual displacement of the peaks of the $Pr_2Fe_{14}B$ phase to the left, indicating the expansion of the unit cell. This is an indicative that all $Pr_2Fe_{14}B$ phase became $Pr_2Fe_{14}BH_x$ (the value of x depends on the temperature and pressure). The alloy shortly exposed to H_2 for 60 and 90 s was unsuitable to be used in the production of sintered magnets. Due to the limited time of reaction, coarse material was found after milling.

Table 2 shows the magnetic properties, $\langle \cos \Theta \rangle$, the MGS and σ of the $Pr_{16}Fe_{76}B_8$ magnets produced employing a hydrogenation time of 120 s and milled for 4.5 and 5.4 ks. Superior milling times make powder extremely difficult due its pyrophoric character. The best overall magnetic properties and alignment degree have been obtained with $Pr_{16}Fe_{76}B_8$ magnets in which the alloy was milled for 5.4 ks. The improvement in $\langle \cos \Theta \rangle$, B_r and $(BH)_{max}$ has been attributed to the larger magnetocrystalline anisotropy of the $Pr_2Fe_{14}B$ phase grains. The decrease of the content of hydrogen in the decrepitated alloy raises the torque of the particles during magnetic orientation. The reduction in grain size provided a higher intrinsic coercivity. The coercivity found in the magnets produced by using this particular processing condition is higher than that found in $Pr_{16}Fe_{76}B_8$ magnets prepared using RBM, which requires milling between 64.8 and 72.0 ks [3,5].

The structure of the shortly hydrogenated $Pr_{16}Fe_{76}B_8$ alloy milled for 5.4 ks is shown in Fig. 4. This structure is similar in shape to that shown in Fig. 2(b). The grain size (3.49 μm) of this magnet is 13% larger than that obtained for the sample prepared using the standard HD process

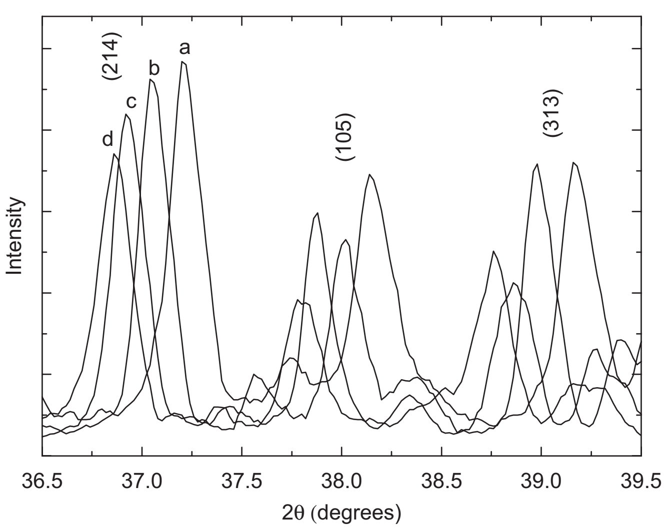

Fig. 3. XRD patterns of the $Pr_{16}Fe_{76}B_8$ alloy: (a) in as-cast state (b) hydrogenated for 60 s, (c) hydrogenated for 120 s and (d) hydrogenated for 3.6 ks.

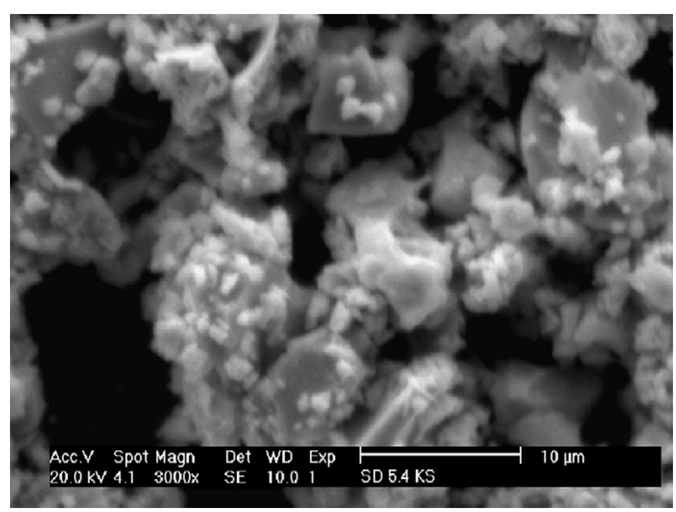

Fig. 4. SEM micrograph of the $Pr_{16}Fe_{76}B_8$ alloy exposed to H_2 for 120 s and milled for 5.4 ks.

Table 2
Properties of magnets produced using a short hydrogenation time (120 s)

Milling time (ks)	B_r (±2%) (mT)	$\mu_{0i}H_c$ (±2%) (mT)	$(BH)_{max}$ (±2%) (kJ m^{-3})	$\langle \cos \Theta \rangle$ (±2%)	MGS (μm)	σ (μm)
4.5	1100	1460	228	0.78	4.06	1.62
5.4	1140	1440	250	0.88	3.49	1.41

(2.95 μm). This is ascribed to lesser content of hydrogen in the decrepitated alloy. The standard deviation of the MGS is 12% smaller than the previous case, showing that long milling times promote a better particle size distribution.

4. Conclusion

In this study, sintered $Pr_{16}Fe_{76}B_8$ permanent magnets with $B_r = 1140 \pm 25\,mT$, $\mu_{0i}H_c = 1440 \pm 30\,mT$, $(BH)_{max} = 250 \pm 5\,kJ\,m^{-3}$ and $\langle \cos \Theta \rangle = 0.88 \pm 0.02$ were produced by decrepitating the cast alloy for 120 s and planetary ball milling for 5.4 ks. These values are similar or even superior (as coercivity) to those obtained in HD sintered $Pr_{16}Fe_{76}B_8$ magnets produced using RBM. A very convenient reduction in the whole processing time of about two-thirds has been achieved by employing high-energy milling.

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