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A Study of Pr-Fe-B Magnets Produced by a Low-Cost Powder Method and the Hydrogen Decrepitation Process

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Sintered Pr-based magnets were produced using a new laboratory technique for powder handling. Unlike the conventional procedure for preparing sintered permanent magnets in the laboratory, the powder technique used in this investigation does not require a glove box. The effects of processing parameters on the magnetic properties of Pr-based sintered magnets prepared using the hydrogen decrepitation process have been studied. Specifically, the effects of sintering temperature and milling time for processing $Pr_{16}Fe_{76}B_8$ magnets have been investigated. $Pr_{16}Fe_{76}B_8$ magnets with the best magnetic properties were sintered between 1015 °C to 1075 °C.

Keywords*: Pr-alloys, magnetic materials, hydrides, magnetic properties*

1. Introduction

The powder transfer technique developed recently in this laboratory was quite satisfactory for preparing neodymium-based sintered magnets[1]. Hydrogen decrepitated (HD) sintered magnets produced using this new technique exhibited magnetic behavior similar to magnets prepared using standard procedures[2,3]. Furthermore, the magnetic properties of the sintered magnets prepared by this low-cost method were equivalent to those of commercial magnets with identical composition[4]. The Pr-based magnets studied previously used the hydrogen decrepitation process followed by roll milling and the powder was handled in a glove box with less than 30 ppm oxygen[5-11]. In this investigation, this new technique has been used for producing praseodymium-based magnets in the laboratory without a glove box. Hydrogen decrepitated $Pr_{16}Fe_{76}B_8$ sintered magnets were produced by this low-cost technique using various milling times. The optimum sintering temperature for this material has also been determined. Hydrogen decrepitated $Pr_{17}Fe_{79}B_4$ sintered magnets have been included in this work for comparison.

2. Experimental Procedure

Commercially available $Pr_{16}Fe_{76}B_8$ and $Pr_{17}Fe_{79}B_4$ alloys were processed in the as-received condition, that is, in the conventional cast ingot state. To prepare Pr-type magnets via the HD process, the following procedure was used[5-8]. Small pieces of the bulk ingot were placed in a stainless steel hydrogenation vessel that was then evacuated to backing-pump pressure. Hydrogen was introduced to a pressure of 1 bar, which resulted in decrepitation of the bulk material. This material was then transferred to a ball milling container which was filled with cyclohexane and roll milled for several hours (9-36 hours). The milling container was then coupled to the transfer system[1]. Figure 1 shows schematically the system used to attain backing-pump vacuum for removal of cyclohexane and for admission of gas for powder transfer. The milled hydride powder was then dried in vacuum for 1 hour and transferred under a N_2 atmosphere to a small cylindrical rubber tube. Powder transfer was carried out while the walls of the system and the isostatic tube were constantly tapped. This system also had a 100 mesh sieve which retained the milling balls and any coarse particles. The outer surface of the isostatic rubber tube was covered with a thin transparent flexible bag for added protection during and after transfer. The fine powder was aligned by pulsing three times in a 6 T magnetic field, pressed isostatically at a pressure of 200 MPa and then vacuum sintered for 1h at 1015-1060 °C, followed by cooling in the furnace. Magnetic characterization of the HD-sintered magnets was carried out using a permaemeter. Measurements were carried out after saturation in a pulsed field of 6 T. The density of the magnets was measured using a liquid displacement system.

3. Results and Discussion

The effects of milling time on the magnetic properties are shown in Figures 2-5 (all sintered at 1060 °C). The most striking feature in these graphs is the variation in remanence (B_r) and inductive coercivity ($_bH_c$) with milling time for both alloys. $Pr_{16}Fe_{76}B_8$ sintered permanent magnets made with powder milled for 18 hours exhibits an increase in remanence. Similarly, the $Pr_{17}Fe_{79}B_4$ HD sintered magnets also exhibit an increase in remanence, but only in those made from powders milled for significantly longer times. In both cases the remanence deteriorats when the powder is milled for prolonged times. The former shows a good value of remanence, 1.19 T and the latter, only 1.14 T. These values are consistent with the higher iron and reduced boron content in the $Pr_{17}Fe_{79}B_4$ alloy.

$Pr_{16}Fe_{76}B_8$ sintered permanent magnets made with powders milled for 18 and 27 hours exhibit a substantial increase in intrinsic coercivity ($_iH_c$). The best value of intrinsic coercivity can be observed in magnets made with powders milled for 27 hours, reaching almost 1.5 T. In a similar manner, the $Pr_{17}Fe_{79}B_4$ HD sintered magnets also exhibit an increase in intrinsic coercivity, but to a lesser extent, if prepared with powders milled for significantly longer milling times. In both cases this magnetic property deteriorates with powders milled for prolonged times. The former alloy showed a reasonable squareness factor (SF) value, of 0.78 and the latter, only 0.61. These values are also consistent with the higher amount of free iron and reduced amount of boron found in the $Pr_{17}Fe_{79}B_4$ alloy. A summary of the magnetic properties and densities (ρ) of the magnets prepared with these two alloys is given in Tables 1 and 2.

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The effect of sintering temperature on the magnetic properties of the $Pr_{16}Fe_{76}B_8$ HD magnets is shown in Figures 6 and 7. $Pr_{16}Fe_{76}B_8$ sintered permanent magnets exhibit a marked increase in remanence when sintered at 1030 °C. Similarly, $Pr_{16}Fe_{76}B_8$ HD sintered magnets also exhibit an increase in the intrinsic coercivity, but when sintered

Table 1. Magnetic properties and density of $Pr_{16}Fe_{76}B_8$ HD magnets prepared with powders milled for various times and sintered at 1060 °C (error: ± 2%).

Milling time (h)	B_r (T)	$\mu_{o\,i}H_c$ (T)	$\mu_{o\,b}H_c$ (T)	$(BH)_{max}$ (kJ/m^3)	SF (ratio)	ρ (g/cm^3)
9	1.02	1.00	0.79	197	0.64	7.28
18	1.19	1.44	1.08	280	0.78	7.33
27	0.95	1.50	0.85	158	0.63	7.37
36	0.92	1.37	0.81	151	0.53	7.32

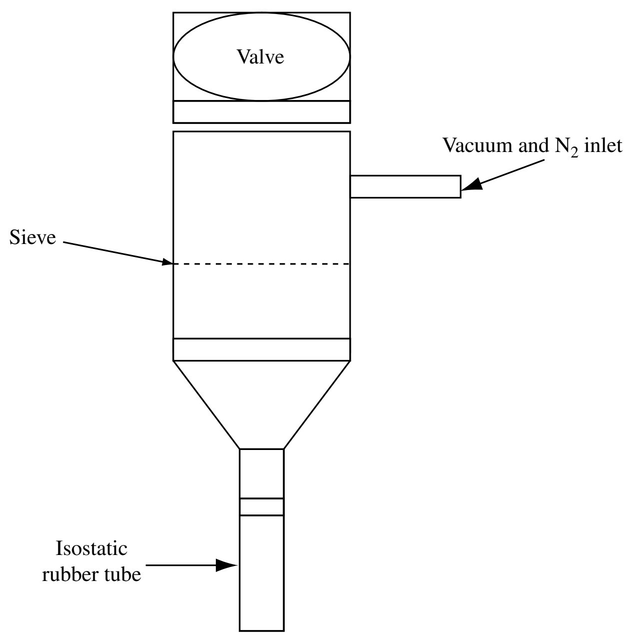

Figure 1. Schematic drawing of the powder transfer apparatus.

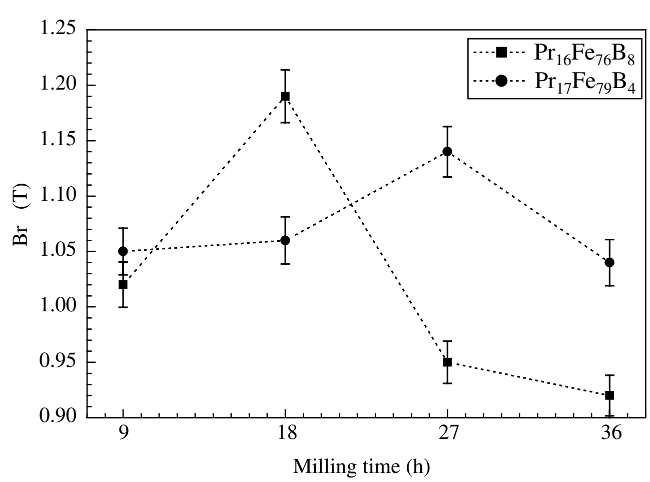

Figure 2. Remanence of $Pr_{16}Fe_{76}B_8$ and $Pr_{17}Fe_{79}B_4$ HD magnets, as a function of milling time.

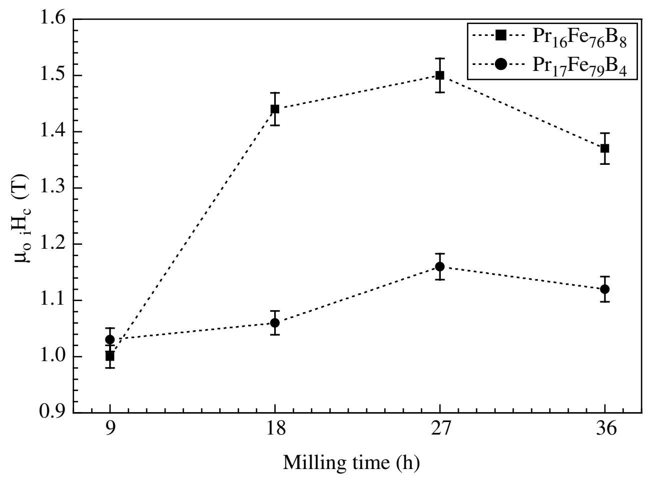

Figure 3. Intrinsic coercivity of $Pr_{16}Fe_{76}B_8$ and $Pr_{17}Fe_{79}B_4$ HD magnets, as a function of milling time.

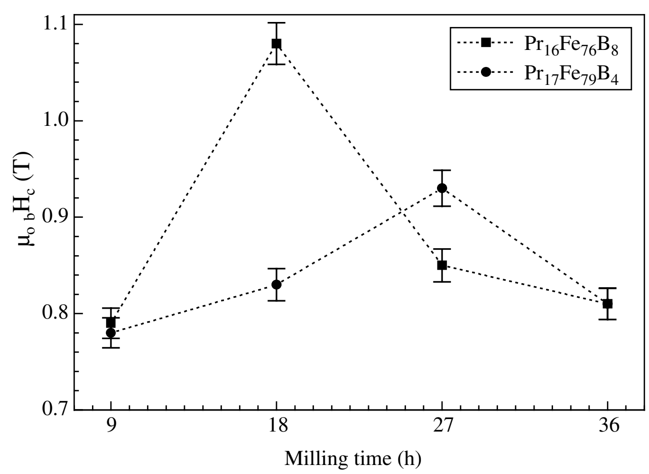

Figure 4. Inductive coercivity of $Pr_{16}Fe_{76}B_8$ and $Pr_{17}Fe_{79}B_4$ HD magnets, as a function of milling time.

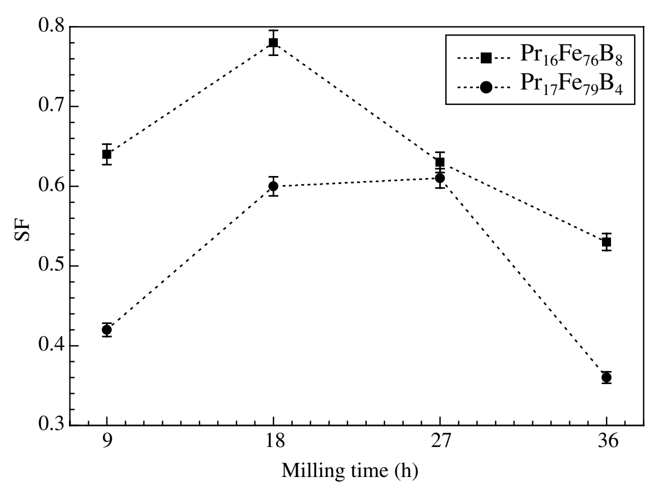

Figure 5. Squareness factor *vs.* of $Pr_{16}Fe_{76}B_8$ and $Pr_{17}Fe_{79}B_4$ HD magnets, as a function of milling time.

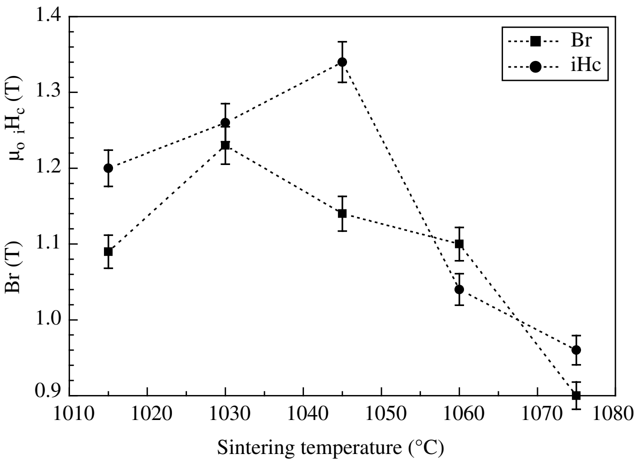

Figure 6. Remanence and energy product *vs.* sintering temperature for $Pr_{16}Fe_{76}B_8$ HD magnets prepared using a hydride powder milled for 20 hours.

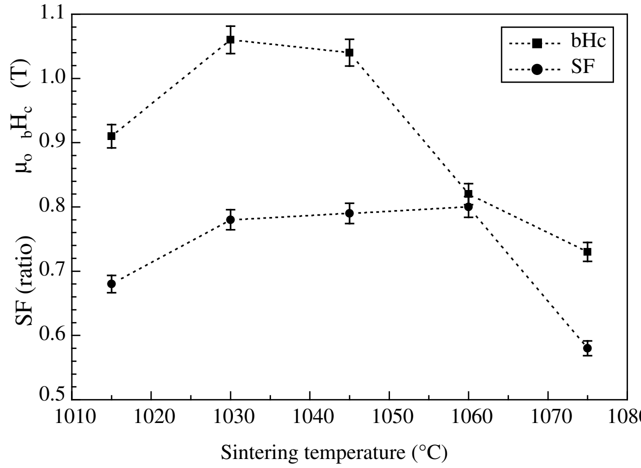

Figure 7. Inductive coercivity and squareness factor *vs.* sintering temperature for $Pr_{16}Fe_{76}B_8$ HD magnets prepared using a hydride powder milled for 20 hours.

Table 2. Magnetic properties and density of $Pr_{17}Fe_{79}B_4$ HD magnets prepared using powders milled for various times and sintered at 1060 °C (error: ± 2%).

Milling time (h)	B_r (T)	$\mu_{o\,i}H_c$ (T)	$\mu_{o\,b}H_c$ (T)	$(BH)_{max}$ (kJ/m³)	SF (ratio)	ρ (g/cm³)
9	1.05	1.03	0.78	167	0.42	7.15
18	1.06	1.06	0.83	197	0.60	7.33
27	1.14	1.16	0.93	217	0.61	7.35
36	1.04	1.12	0.81	175	0.36	7.37

Table 3. Magnetic properties of $Pr_{16}Fe_{76}B_8$ HD magnets prepared with a powder milled for 20 hours and sintered at various temperatures. (error: ± 2%).

Sintering temperature (°C)	B_r (T)	$\mu_{o\,i}H_c$ (T)	$\mu_{o\,b}H_c$ (T)	$(BH)_{max}$ (kJ/m³)	SF (ratio)	ρ (g/cm³)
1015	1.09	1.20	0.91	213	0.68	7.26
1030	1.23	1.26	1.06	285	0.78	7.50
1045	1.14	1.34	1.04	240	0.79	7.49
1060	1.10	1.04	0.82	215	0.80	7.53
1075	0.90	0.96	0.73	172	0.58	7.48

at a slightly higher temperature (1045 °C). Both these properties deteriorate in magnets sintered at higher temperatures. Inductive coercivity increases steadily with sintering temperature in the HD powder, whereas the squareness factor reaches a peak at 1030 °C and, following the general behavior, deteriorates at higher temperatures. A summary of magnetic properties and density is given in Table 3.

4. Conclusions

Hydrogen decrepitated sintered permanent magnets have been successfully produced by the new low-cost laboratory technique. Sintered HD magnets based on $Pr_{16}Fe_{76}B_8$ exhibited a peak in remanence and coercivity when prepared with powders milled for shorter times compared to $Pr_{17}Fe_{79}B_4$ magnets. Optimum milling time for the powders of the former was 18 hours and for the latter, 27 hours. A permanent magnet produced from a powder milled for 20 hours and sintered at 1030 °C exhibited remanence of 1.23 T and intrinsic coercivity of 1.26 T.

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