Magnetic properties of bulk polycrystalline $Pr_{1-x}Nd_x$ ($x = 0$, 0.8)FeB permanent magnets*

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For spin reorientation (SRT), the applications of sintering NdFeB permanent magnets are limited at low temperature. The sintering PrFeB permanent magnet (PM) presents no SRT and shows excellent magnetic properties at low temperature. The magnetic properties of bulk polycrystalline sintering $Pr_{1-x}Nd_xFeB$ ($x = 0$ and 0.8 correspond to P42H and N50M respectively) are studied in this paper. The results show that magnetic properties and stability of N50M are better than those of 42H at room temperature. With the decrease of temperature, the parameters of B_r, H_{cb}, and H_{ci} of P42H present a nearly linear increasing trend; B_r and H_{cb} of N50M first increase and then decline, H_{ci} presents an increasing trend. At 77 K, B_r, H_{ci}, and J_r of P42H are increased by 18.7%, 308%, and 17.1% respectively over than those at 300 K; at 120 K, B_r, H_{ci}, and J_r of N50M are increased by about 16.19%, 245%, and 12.6% respectively over than those at 300 K. The magnetic properties of P42H are better than those of N50M at low temperature. The sintering PrFeB is the preferred PM in various low-temperature devices.

Keywords: PrFeB, NdFeB, bulk polycrystalline, magnetic properties

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1. Introduction

Due to ultrahigh magnetic energy and cost advantages, NdFeB permanent magnets (PMs) have broad applications in various industries.[1] In the last century, magnetic properties of single crystal NdFeB have been investigated at low temperatures. In recent years, magnetic properties of commercial bulk polycrystalline NdFeB have also been studied in some fields (such as synchrotron radiation, space detectors, and industries),[2–5] and the results show that NdFeB presents spin reorientation (SRT) at 135 K,[6] which will restrict its applications at low temperatures.

Magnetic structure and theoretical magnetic properties of PrFeB[7,8] are close to those of NdFeB. At present, there are a few of reports on magnetic properties of bulk polycrystalline sintering PrFeB. In 2013, the Shanghai Institute of Applied Physics and Zhejiang Innuovo Magnet Co., Ltd jointly developed sintering $Pr_{1-x}Nd_xFeB$ ($x = 0$, 0.8, correspond to P42H and N50M respectively) and their magnetic properties have been studied, which are described in this paper.

2. Magnetic properties at room temperature

N50M and P42H were made by powder metallurgy process, and their B & J–H curves and magnetic parameters at room temperature are shown in Fig. 1 and Table 1 respectively.

Figure 1 shows that B_r and H_{cb} of N50M are better than those of P42H, H_{ci} is less than that of P42H, and the values of reply permeability μ_r of N50M and P42H are 1.01 and 1.03 respectively. The difference between magnetic properties is closely related to the composition and preparation technology of alloys. After nearly three-decades of development, the design and preparation methods of NdFeB have become relatively mature, although Pr replaces 20% of Nd for using NdFeB preparation technology, N50M shows higher magnetic properties; PrFeB powder is more easily oxidized, and the preparation technology is not perfect. For fabricating the bulk polycrystalline magnets with high magnetic properties, the component design and preparation technique need to be investigated.

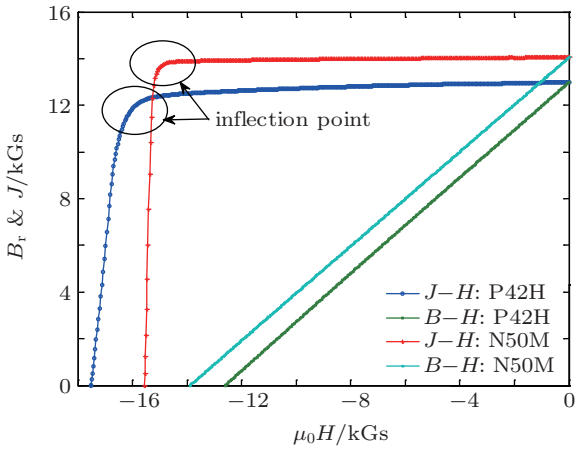


Fig. 1. (color online) B–H and J–H curves of N50M and P42H, 1 Gs = 10^{-4} T.

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From Table 1 it follows that the square Q (H_k/H_{ci} at infection point), B_r temperature coefficient α, and H_{ci} temperature coefficient β of N50M are better than those of P42H, the Curie point T_c of N50M is greater than that of P42H. These indicate that magnetic field stability of PrFeB is inferior to that of NdFeB magnet at room temperature, which is one of main reasons why the PrFeB is rarely used commercially. If PrFeB can have applications, it needs to improve H_{ci} and to maintain good Q and to reduce α, β, but this will result in the reduction of B_r.

Table 1. Magnetic parameters of N50M and P42H at room temperature.

Brand	B_r/T	H_{cb}/(kA/m)	H_{ci}/(kA/m)	Q/%	α	β	T_c/°C
N50M	1.41	1108	1220	98.0	0.10	0.60	313
P42H	1.30	1004	1400	91.9	0.12	0.90	280

3. Magnetic properties at low temperature

3.1. *J–H* behavior

The *M–H* behavior (parallel to orientation) was tested by using the physical property measurement system (PPMS) at Shanghai Jiaotong University. The sample PMs with size 2.0 mm×2.0 mm×2.5 mm (orientation 2.5 mm) were not magnetized. Figure 2 show *J–H* ($J = \mu_0 M$) in the second quadrant, from which we can see that N50M shows magnetization reversal (Mag. Rev.) only at 300 K and 150 K, but show SRT and Mag. Rev. at 80 K simultaneously. P42H only presents Mag. Rev. and no SRT; but in the first quadrant, P42H presents Mag. Rev., which is related to partial oxidation of the magnet, resulting in Pr_2Fe_{17} phase. Compared with $Pr_2Fe_{14}B$, Nd_2Fe_{17} with low anisotropy field H_A may be first formed in Mag. Rev. domains in the first quadrant. The Mag. Rev. is also closely related to the shape and microstructure inhomogeneity of PMs.

From formula $B = M + H$, the obtained *B–H* curve in the second quadrant is shown in Fig. 3.

From Fig. 3, at 80 K, 150 K, and 300 K, the values of H_{cb} for N50M are 1020 kA/m, 1084 kA/m, and 757 kA/m respectively; at 100 K, 200 K, and 300 K, the values of H_{cb} for P42H are 1188 kA/m, 1100 kA/m, and 741 kA/m respectively. The *B–H* linearity of P42H was significantly improved by reducing temperature. At 100 K and 200 K, from equation $\mu_r = B_r/H_{cb}$, the values of reply permeability μ_r corresponding to 1.02 and 1.05 are obtained respectively. But for N50M, at 80 K and 150 K, the values of μ_r are 1.25 and 1.21 respectively. The mean linearity of N50M is inferior to that of P42H at low temperature. At 300 K, *B–H* curves of the two magnets become bent when H is close to −0.9 T, but at low temperatures (80 K, 150 K, 100 K, and 200 K), *B–H* curves of N50M and P42H each show a nearly linear characteristic: this phenomenon is closely related to H_{ci} and slim sample PM shape.

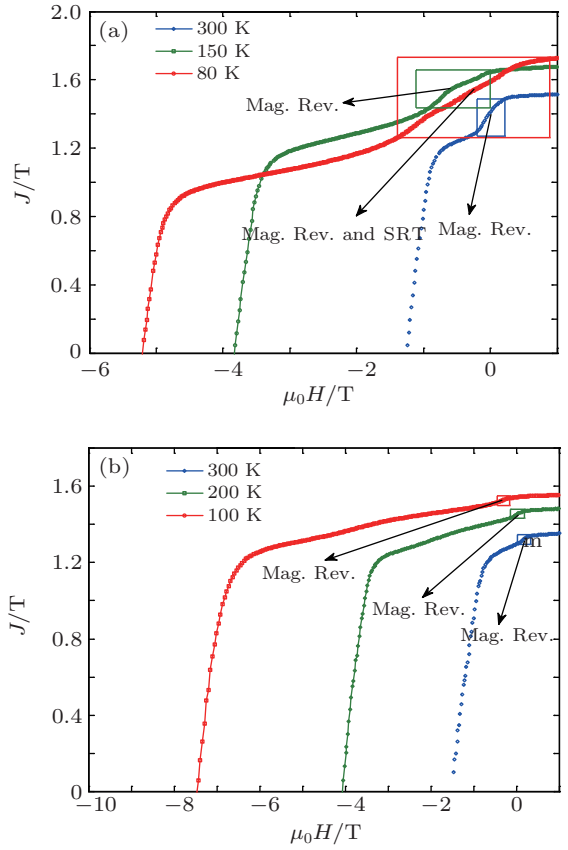


Fig. 2. (color online) *J–H* curves of (a) N50M and (b) P42H.

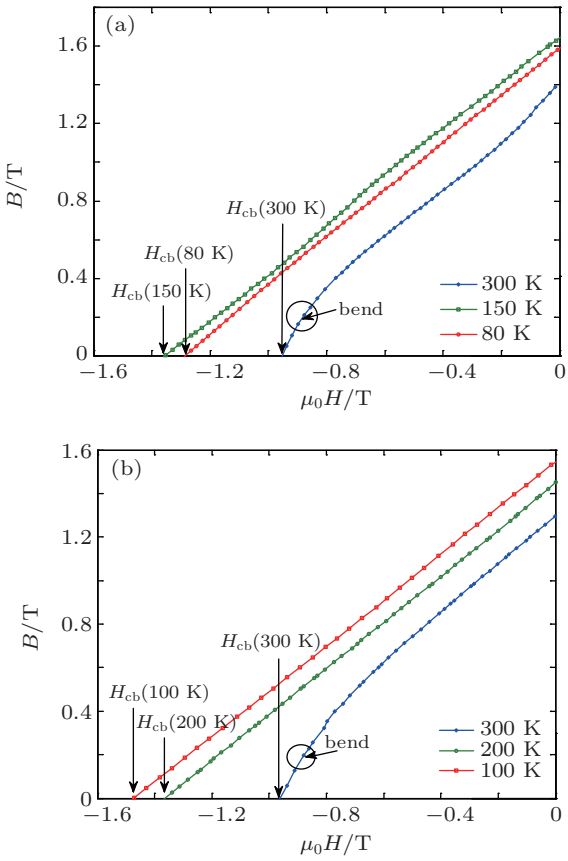


Fig. 3. (color online) *B–H* curves of (a) N50M and (b) P42H.

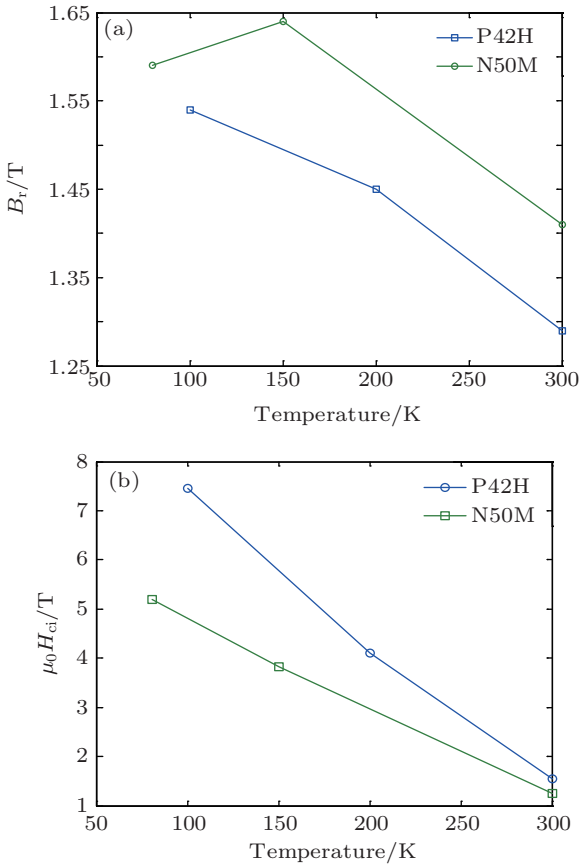


Fig. 4. (color online) (a) B_r and (b) H_{ci} at low temperatures.

Intersection points of J–H curve and axis are B_r and H_{ci} respectively, as shown in Fig. 4. With the decrease of temperature, B_r of N50M first increases and then decreases, and B_r of P42H shows an increasing trend, the values of H_{ci} of N50M and P42H increase always. It can be calculated that at 77 K, the values of B_r and H_{ci} of P42H are increased by 18.7% and 308% compared with those at 300 K respectively, and at 120 K, the values of B_r and H_{ci} of N50M are increased by 16.19% and 245% compared with those at 300 K.

3.2. Orientation degree

For NdFeB or PrFeB, the orientation degree represents the anisotropy degree, and it can be assessed by formula of B_r:[9]

$$B_r = A(1-\beta)D(\cos\theta)J_s, \tag{1}$$

where $J_s = \mu_0 M_s$, $B_r = \mu_0 M_r$, A is the volume fraction of forward domain, β is the volume fraction of non-magnetic phase, D is the relative density, $\cos\theta$ is the orientation degree (θ see Fig. 5), J_s is the saturated magnetic polarization. The values of A for N50M and P42H are both about 1.0, the values of β are both about 0.02, the values of D are about 0.98 and 0.99 respectively, B_r and M_s can be obtained from the J–H curve. The $\cos\theta$ can be calculated from formula (1), and the results are given in Table 2. We can see that $\cos\theta$ of P42H increases gradually and $\cos\theta$ of N50M first increases and then decreases with the decrease of temperature.

Table 2. Orientation degrees of N50M and P42H.

Temperature/K	N50M			Temperature/K	P42H		
	B_r/T	M_sT	$\cos\theta$		B_r/T	M_s/T	$\cos\theta$
300 K	1.406	1.552	0.941	300 K	1.30	1.419	0.951
150 K	1.643	1.740	0.981	200 K	1.452	1.540	0.979
80 K	1.590	1.776	0.930	100 K	1.544	1.597	1.000

3.3. J_r–T behavior

The J_r–T ($J_r = \mu_0 M_r$) behaviors are also tested by PPMS (as shown in Fig. 5).

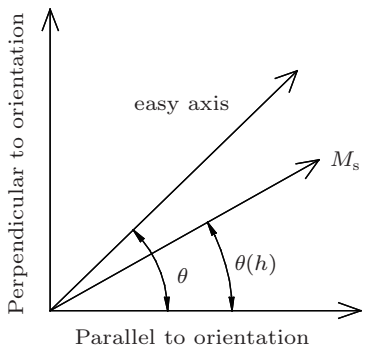


Fig. 5. Easy axis and orientation axis.

The rectangular sample PMs with the following size: 2.0 mm×2.0 mm×2.5 mm (orientation 2.5 mm) are saturation-magnetized. From Fig. 6(a), one can see the parallel orientation, owing to the SRT. The N50M shows residual magnetic polarization J_r peaking at 120 K, showing that the value of J_r is increased by 12.6% compared with that at 300 K. With the decrease of temperature, the value of J_r of P42H is nearly linearly increased, and at 77 K the value of J_r is increased approximately by 17.1% compared with that at 300 K. From Fig. 6(b), one can see the perpendicular orientation. The values of J_r of N50M and P42H show "jumps" at 235 K and 225 K respectively, then present significant jitters in a very wide temperature range. For high precision PM devices, if they are to be used in that temperature range, much

attention must be paid to this phenomenon.

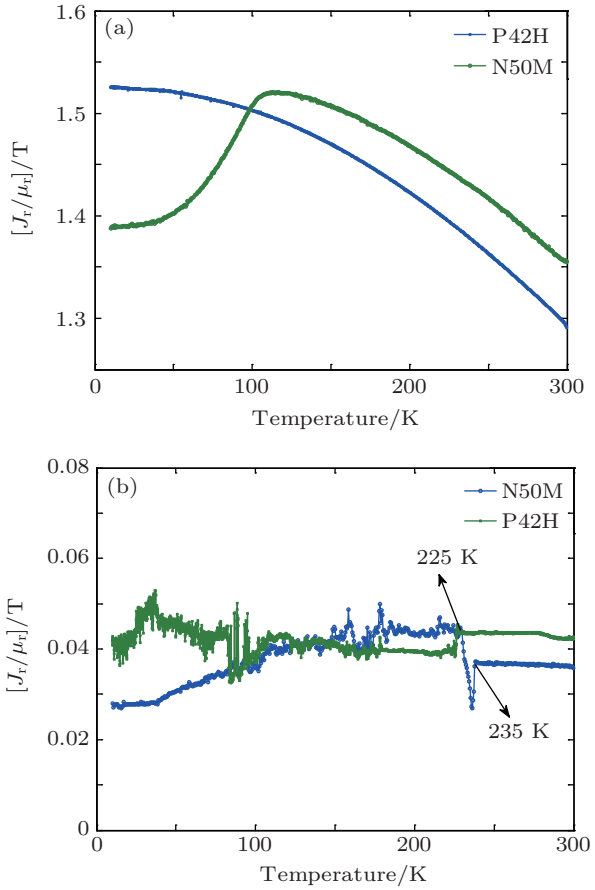


Fig. 6. (color online) J_r–T behaviors at (a) parallel orientation and (b) perpendicular orientation.

4. Discussion

When a single crystal $R_2Fe_{14}B$ magnet is magnetized along the easy axis direction, $M_r = M_s$ from $J_s = \mu_0 M_s$ and $B_r = \mu_0 M_r$, then the limit value of B_r is J_s, and the limit value of H_{ci} for single crystal can be calculated by the following formula:

$$H_{ci} = 2(K_1 + K_2)/\mu_0 M_s, \tag{2}$$

where K_1 and K_2 are the anisotropy constants. For a bulk polycrystalline $R_2Fe_{14}B$ magnet, the limit value of B_r is the same as that of single crystal, but the limit value of H_{ci} is less than that of single crystal. These values can be seen in Table 3.

Table 3. Limit values of B_r and H_{ci} for $R_2Fe_{14}B$ (B_r and H_{ci} are in units of T and kA/m, respectively, and K_1 and K_2 are in units of MJ/m^3).[10]

$R_2Fe_{14}B(R=)$		293 K				4.2 K			
		B_r	K_1	K_2	H_{ci}	B_r	K_1	K_2	H_{ci}
Single	Nd	1.60	5.0	0.66	7077	1.86	–16	28	16.2
crystal	Pr	1.56	5.6		7173	1.84	24		32.7
Poly-	Nd	1.60			5580				
crystalline	Pr	1.56			5898				

The value of B_r of N50M is 1.41 T, and there is a small gap compared with the limit value 1.60 at room temperature. The value of H_{ci} is 1240 kA/m, and there is a bigger gap compared with the limit value 5580 kA/m. The values of B_r and H_{ci} of P42H are 1.30 T and 1400 kA/m at room temperature, and there are bigger gaps compared with the limit value 1.56 T and 5898 kA/m respectively. Being relative to B_r, H_{ci} of bulk polycrystalline PM is very sensitive to magnetic structure and technological factors that greatly affect it. The magnetic phase of P42H is $Pr_2Fe_{14}B$, and K_1 has a positive value always and has no SRT, and its magnetic properties are lower than those of N50M at room temperature, but at low temperatures, the values of B_r and H_{ci} of P42H are greater than those of N50M. The value of K_1 of NdFeB would change from positive to negative value at about 130 K. the NdFeB single crystal begins to show the SRT at about 135 K, but bulk polycrystalline N50M magnetic phase has some $Pr_2Fe_{14}B$ and $(NdPr)_2Fe_{14}B$, which induces the starting point of SRT to move down to 120 K nearby.

5. Conclusion

At present, the available high-performance rare earth PMs are NdFeB, Sm_2Co_{17}, and PrFeB. Figure 7 shows typical domestic PMs brand, from which it can be seen that Sm_2Co_{17} has a good stability and wide applications in In-vacuum undulators,[11] but its magnetic properties are lower. NdFeB with higher H_{ci} such as N33VH, shows small B_r at low temperature; NdFeB with higher B_r and lower H_{ci} such as N48H, may show higher B_r and H_{ci} at low temperature simultaneously, which is suited to some applications at low temperatures, but SRT and poor B–H linearity will bring some challenges to the development of cryogenic PM devices. PrFeB shows excellent cryogenic magnetic properties and good B–H linearity and so on, which make it more suitable for a variety of cryogenic PM devices, if magnetic properties of PrFeB can reach the current level of commercial NdFeB at room temperature, B_r and H_{ci} of the PM will be very considerable at low temperatures.

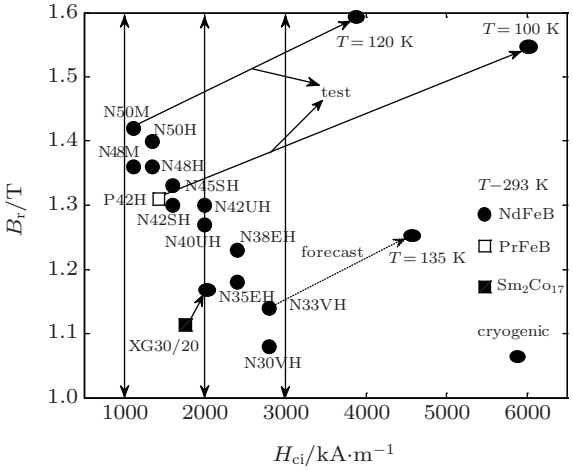


Fig. 7. Magnetic properties of typical domestic PMs.

If preparation technology and stability of PrFeB can be solved at room temperature, and its magnetic properties are

then gradually increased, the PMs have broad application prospects in low-temperature devices such as cryogenic undulators of synchronous radiation, free electron lasers, and space instrumentation, and so on.

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