MAGNETIC PROPERTIES OF SINTERED Pr-Fe-B-Cu HD MAGNETS

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ABSTRACT

Sintered permanent magnets of PrFeBCu-type have been prepared using the hydrogen decrepitation (HD) process. The relationships between processing conditions and magnetic properties have been studied. Post sintering heat treatment by annealing the $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ as-sintered samples at 1000°C has been shown to be beneficial for increasing the intrinsic coercivity. For a specific processing condition the intrinsic coercivity double with this high temperature heat treatment. The increase in the coercivity has been partially attributed to the diminution in the amount of a 2/17-type phase. The possible role of this phase on the coercivity behaviour of sintered RE-Fe-B-Cu (RE = Nd and Pr) permanent magnets has also been discussed.

Key Words: Permanent Magnets; Hydrogen Decrepitation Process;Rare Earth-Fe-B Magnets

53º Congresso Anual da ABM

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INTRODUCTION

The influence of B content on the magnetic properties of $RE_{17}Fe_{83-x}B_x$ (RE= Ce, Nd and Pr, and x=1-7) sintered permanent magnets has been reported[1]. In this investigation, the sintered magnets were prepared by conventional powder metallurgy method using a standard milling step and with no post sintering heat treatments applied to the magnets. Recently, it was shown that magnets based on the compositions $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ and $Pr_{16.9}Fe_{79.1}B_4$, produced using the hydrogen decrepitation process, achieved high coercivity after a post sintering heat treatment[2,3]. In the present study, HD sintered permanent magnets of a $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ alloy were prepared using a wide range of milling times. In addition, a post sintering high temperature heat treatment was applied to the magnets and the effects of this treatment studied.

It has been shown that Nd-Fe-B magnets with boron content less than 5 at.% have lower magnetic properties than the counterpart Pr-based magnets with similar compositions[1]. This has been attributed to the higher magnetocrystalline anisotropy field (H_A) of the $Pr_2Fe_{14}B$ matrix phase and the low spontaneous magnetization (M_S) of the Pr_2Fe_{17} phase . In this paper, the possible role of the 2:17 type phase on the coercivity behaviour of sintered Pr-Fe-B-Cu magnets has also been discussed.

EXPERIMENTAL

The bulk alloy was produced by induction-melting the constituents in vacuum and then casting in rectangular (20x10x3 cm^3) water cooled copper moulds (composition in Table 1). The microstructural observations and microanalysis were carried out on the $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ alloy using an optical microscope and a JEOL 840A scanning electron microscope, respectively. The analyses of the matrix phase, grain boundaries, Pr-rich and free iron were carried out by spot analysis on two distinct areas each. A general area, using a 200 mm spot, has also been analysed.

Table 1. Nominal composition of the as-cast alloy.

Atomic %	Wt %			
	Pr	Fe	B	Cu
$Pr_{17}Fe_{76.5}B_5Cu_{1.5}$	35.0	Bal.	0.80	1.30

In order to produce the magnets via the HD process[4], the following procedure was adopted. Small pieces of the bulk ingot were placed in a hydrogenation vessel. This was then evacuated to backing-pump pressure and hydrogen introduced to a pressure of 10 bars. The decrepitated material was then transferred to a "roller" ball-mill under a protective atmosphere and milled using cyclohexane as the milling medium. The resultant fine powder was then dried and encapsulated in a small cylindrical rubber bag, pulsed at a magnetic field and isostatically pressed. The resulting green compacts were then vacuum sintered at 1060∞C for 1 hour and furnace cooled. The sintered magnets were then pulsed in a high magnetic field and their second quadrant demagnetization curves determined. The as-sintered magnets then received a post sintering heat treatment by annealing under vacuum at 1000∞C for 24 hours (furnace cooled) and their magnetic properties re-measured.

RESULTS AND DISCUSSION

Figure 1 shows the microstructure of the alloy in the cast condition. The microstructure of this alloy consists of a $Pr_2Fe_{14}B$ matrix phase, Pr-rich grain boundary regions, B-rich phase ($Pr_{1+e}Fe_4B_4$) and free iron (a Fe) inside the matrix phase. The chemical compositions of the main phases present in the microstructures of the $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ alloy are given in Table 2. As expected the matrix phase was found to be close to the composition $Pr_{11.8}Fe_{82.4}B_{5.8}$ but some under estimation in the amount of boron was observed for the general area analysis. In the grain boundary regions a eutectic mixture is also visible. It has been shown that after annealing, the eutectic phase region is more clearly visible and enriched in copper[5]. No clear evidence of 2/17 phase observed in sintered magnets[1,3,6,7] has been found in this cast material.

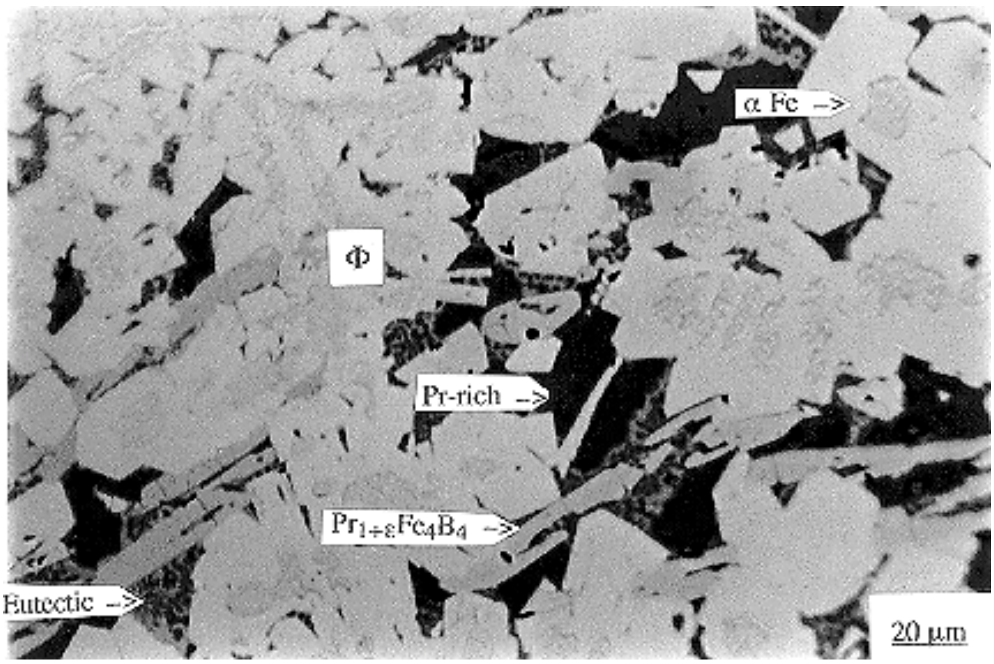

Fig 1 Details of the as-cast microstructure of the $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ alloy (optical).

Table 2. Chemical composition of the phases present in the microstructure of the $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ alloy in the cast state (Error ± 1.0).

Phase/ Area	**Phase** Identification	**Composition at%** Pr	Fe	B	Cu
General Area	Spot = 200 _m	16.6	79.3	1.9	2.2
Matrix _	Gray	12.6	81.4	6.0	---
Primary Pr-rich	Dark	97.4	0.2	0.6	1.8
Eutectic	Light+dark	91.9	1.9	0.3	5.9
α Fe	Darker gray inside _	0.8	99.2	---	---

The effect of milling time and annealing at 1000°C for 24 hours for the $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ magnets are shown in Fig.2, and the demagnetization curve for the best magnet is shown in fig. 3. Some improvement in the magnetic properties has been obtained with the high temperature heat treatment for milling times up to 36 hours. Nd-based magnets with the same composition and heat treatment showed inferior magnetic properties[6]. Better magnetic properties have been achieved using an optimum heat treatment (1100°C for 1 h and 600°C for 1h)[7,8,9].

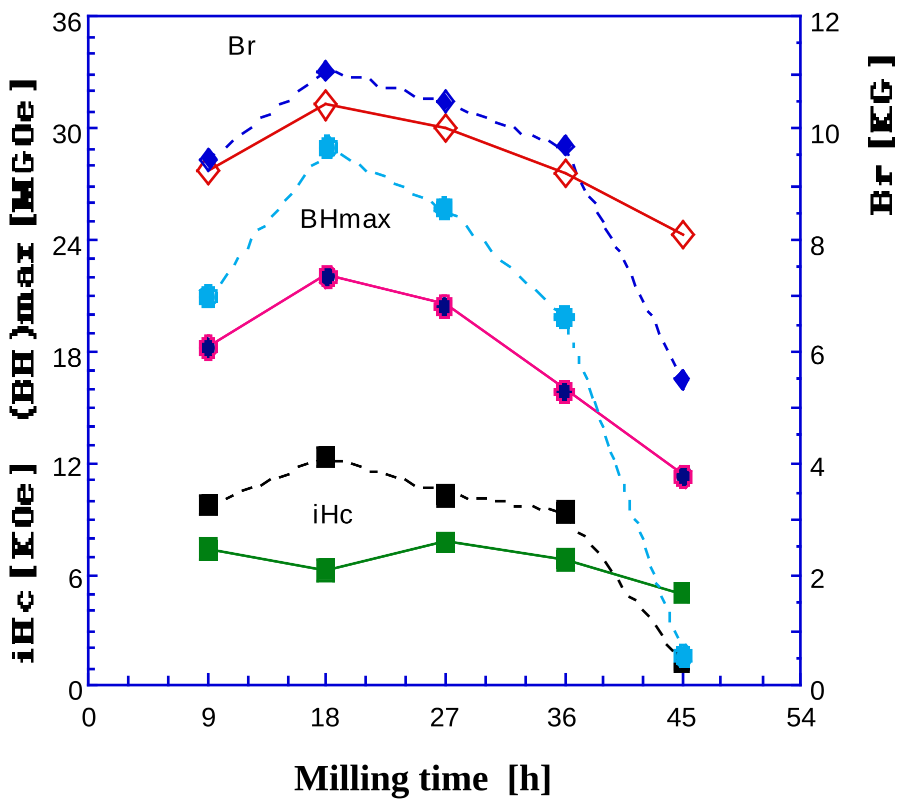

Fig.2 Variation of (BH)max (o) and iHc (■) and Br (♉) with the milling time for slow cooled HD magnets of $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ alloy (Black/Dashed : annealed 1000°C, 24 hours).

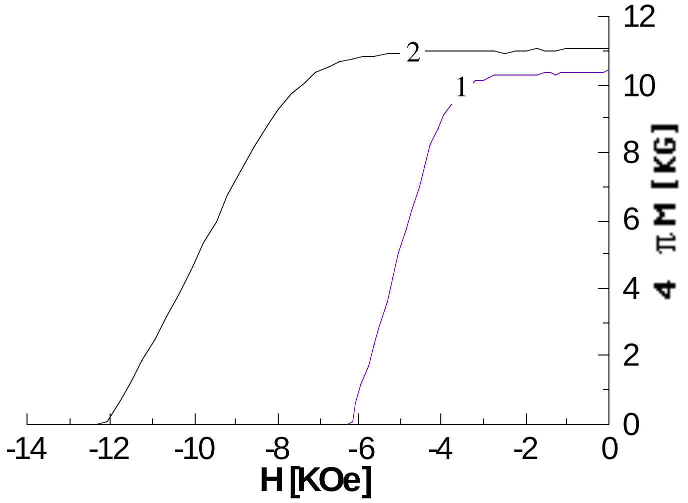

Fig. 3 The demagnetization curves for slow cooled $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ magnets, before (1) and after annealing (2) (milling time 18 hours).

It has been shown[1] that, in sintered magnets of the $RE_{17}Fe_{83-x}B_x$-type, the magnetically soft RE_2Fe_{17} phase always occurs when x<5, and this phase would act as nucleation centres for magnetization reversal. Previous studies[3,10] have shown the presence of 2/17 phase in HD sintered and hot pressed $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets and that the amount of this phase diminished after heat treatment. The increase in the intrinsic coercivity has been attributed partially to the diminution in the amount of the Pr_2Fe_{17} phase. Similarly a Nd_2Fe_{17} magnetically soft phase has also been found in $Nd_{17}Fe_{76.5}B_5Cu_{1.5}$ HD sintered magnets[6,7,8]. It has also been shown[7,8] that the amount of this phase was reduced with a high temperature heat treatment. The free iron observed in the cast material has been removed completely by powder metallurgy processing. A comparison of $RE_{17}Fe_{76.5}B_5Cu_{1.5}$ sintered magnets is given in Table 3.

Table 3. Magnetic properties of $RE_{17}Fe_{76.5}B_5Cu_{1.5}$ sintered magnets.

ALLOY TYPE	Processing condition	Br KG	iHc KOe	bHc KOe	BH MGO	S.F ratio	dens. g/cc	REF.
$Pr_{17}Fe_{76.5}B_5Cu_{1.5}$	18h As-sintered	10.4	6.4	5.4	22.1	0.64	7.3	This work
	Annealed	11.1	12.4	8.5	29.0	0.60		
$Nd_{17}Fe_{76.5}B_5Cu_{1.5}$	18h As-sintered	10.3	8.4	6.9	23.2	0.64	7.4	6
	Annealed	10.6	10.0	7.5	24.5	0.60		

(Average error: Br : ± 0.1, iHc : ± 0.5, BHmax : ± 0.9)

Neodymium-based sintered magnets show lower coercivities than the Pr-based magnets and this has been attributed to the higher magnetocrystalline anisotropy field (H_A) of the $Pr_2Fe_{14}B$ matrix phase and the low M_S of the Pr_2Fe_{17} phase (boron content < 5 at.%)[1]. The overall picture of the behaviour of the 2/17 phase can be summarized as follows. When a certain demagnetizing field (the nucleation field) is applied to the $Pr_2Fe_{14}B$ grains a reverse domain will nucleate and grow. The total demagnetizing field applied to each $Pr_2Fe_{14}B$ grain will comprise the externally applied field and the field produced by the reversed magnetized neighbouring grains. The Pr_2Fe_{17} phase is ferromagnetically soft at room temperature, and hence will easily reverse to the spontaneous magnetisation of 6.6 kG [1]. As the Ms of Pr_2Fe_{17} is less than that of Nd_2Fe_{17} (10.7 kG[1]), the former contributes less to the demagnetizing field than the latter. Hence a higher external field must be applied to the Pr-based magnet to achieve an equivalent demagnetizing field to the Nd-based magnet. Therefore the Pr-based magnet will have a higher intrinsic coercivity than the Nd-based magnet if the nucleation field is the same for $Pr_2Fe_{14}B$ and $Nd_2Fe_{14}B$ (also assuming that the amount of 2/17 phase is the same in both magnets). In fact, the nucleation field for $Pr_2Fe_{14}B$ is greater than that for $Nd_2Fe_{14}B$ due to the higher anisotropy field (H_A) of $Pr_2Fe_{14}B$. These two effects, the lower Ms of Pr_2Fe_{17} and the higher H_A of $Pr_2Fe_{14}B$, combine to give a higher coercivity in Pr-based magnets compared to Nd-based magnets.

CONCLUSIONS

In general, the magnetic properties of HD sintered permanent magnets of a $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ alloy can be improved by annealing. For a specific processing condition the intrinsic coercivity double with this high temperature heat treatment . The increase in the coercivity has been partially attributed to the diminution in the amount of a 2/17-type phase. The lower Ms of Pr_2Fe_{17} and the higher H_A of $Pr_2Fe_{14}B$ combine to give a higher coercivity in Pr-based magnets compared to Nd-based magnets.

ACKNOWLEDGMENTS

Many thanks are due to the Instituto de Pesquisas Energéticas e Nucleares (IPEN) and Concerted European Action on Magnets (CEAM) for supporting this research programme.

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