STUDY ON Pr-Fe-B-Cu MAGNETS PRODUCED BY UPSET FORGING OF CAST INGOT (01)

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SUMÁRIO:

Ímàs permanentes anisotrópicos de Pr-Fe-B-Cu foram produzidos de liga fundida pelo processo de forjamento à quente. Ímàs baseados na composição de $Pr_{20,5}Fe_{73,8}B_{3,7}Cu_{2,0}$ alcançaram uma coercividade intrinseca maior que 21 KOe (> 1674 KA/m) com tratamentos térmicos após o forjamento. Uma moderada temperatura de forjamento de 750°C foi utilizada no presente trabalho. Os ímàs forjados foram tratados termicamente em 1000°C por 5 horas e em 500°C por 3 horas. Taxas de deformação (ε) de 2,7 X 10 até 3,0 X 10 e reduções na altura de 80 a 90% foram efetuadas no presente trabalho. As perdas de fase rica em praseodímio, durante o forjamento, foram anuladas pelo encapsulamento das amostras em um cilindro de cobre. Uma investigaçào das propriedades magnéticas da liga fundida depois dos tratamentos térmicos, acima especificados, também foram feitos para uma comparaçào de resultados.

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STUDY ON Pr-Fe-B-Cu MAGNETS PRODUCED BY UPSET FORGING OF CAST INGOT

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INTRODUCTION

In privious studies[1-3] the alloys, $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ and $Pr_{15}Fe_{78.5}B_5Cu_{1.5}$ have been hot pressed at around 1000 °C to produce magnets with good magnetic properties. Recently, however, it has been shown that magnets based on the composition $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ produced by upset forging achieve good magnetic properties at lower forging temperatures (900 °C)[4]. In the present work, upset forged $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets have been produced and investigated after forging at a lower temperature of 750 °C. In previous studies[1-4], during hot working in open dies there was a loss of the Pr-rich phase and consequently deviation from the original composition. In this work, a copper cylinder surrounding the cast ingot has been used to reduce this effect by providing partial constraint. a range of strain rates and thickness reductions have been used in order to study their influence on the magnetic properties. A standard post-upset forging heat treatment at two temperatures (1000 °C and 500°C)[4,5] has been applied to all samples. The degree of alignment obtained during the upset forging process has been investigated by comparing the remanence of the forged magnets with that of the as cast alloy after being subject to an identical annealing treatment[5,6].

EXPERIMENTAL

The study of the $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ cast alloy, the details of the preparation of the upset forged magnets and the upset forging equipment have all been described in previous papers[4,5]. The alloy was prepared in a rectangular (10.0X7.5X1.0 cm) water cooled copper mould and several rectangular blocks of cross-section 10X10 mm² and of heights 25 to 50 mm, were cut from the cast ingot, so that the height of the specimen was perpendicular to the cooling direction of the cast ingot. The blocks were embedded in a copper tube by filing-away the sharp corners of the blocks, and the enclosed sample was then upset forged in a similar manner to that reported previously[4], under vaccum, at a temperature of 750°C. A range of strain rates and thickness reductions were employed. A two stage post-upset heat treatment of the forged ingot was carried out in vacuum at 1000°C for 5 hours followed by quenching to room temperature, and then at 500°C for 3 hours and again quenched to room temperature. Suitable samples (8X8X5 mm³) were cut from the upset forged material for permeameter measurements. The microstructure of the upset forged magnets were observed using an optical microscope and a scanning electron microscope (SEM), and the magnetic domain structure of the specimens was investigated using a ferrofluid technique. Density measurements were carried out by a displacement method using a sensitive balance and diethylphthalate. The remanence of the cast magnets, used as a reference for the evaluation of the degree of alignment of the forged magnets, were obtained after a homogenization heat treatment of the cast ingot carried out under vacuum at 1000°C for 10 hours followed by quenching to room temperature.

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RESULTS AND DISCUSSION

Table I shows the magnetic properties found in various randomly cut samples of the as-cast ingot after annealing at 1000°C. Before annealing, the as-cast alloy exhibit very poor magnetic properties. As can be seen, significant differences in intrinsic coercivity are observed for different regions of the ingot. The remanence, however, is less affected. The lower values of intrinsic coercivity found here compared with previous work[4] can be attributed to the different solidification conditions used during ingot preparation. The average remenence (5.64 KG ± 0.05) found in the present work is also lower than that previously reported on directional solidification studies (7.2 KG[6] and 7.1 KG[8]) since those were maximum values (peak) for their particular solidification conditions and also due to small variations in the alloy composition[8]. This average remanence will be used, later in this work, as reference for the estimation of the approximate degree of alignment obtained during upset forging. Cast magnets based on the present alloy have been studied extensively[6,7].

Figure 1 shows the variations of the magnetic properties of upset forged at 750°C and heat treated $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets as a function of the reduction in thickness for a constant strain rate of 8.0 X 10^{-2} s^{-1}. The intrinsic coercivity increases steadily from 80 to 84% of thickness reduction and reaches a maximum value of 21.5 KOe. It then decreases slowly as the thickness reduction is increased further. Both remanence and energy product follow a similar trend with thickness reduction with higher values at around 82%. The average remanence (from 80 to 90%) is 8.32 ± 0.05 KG which compared with the average remanence of the annealed as-cast alloy gives an average increase on the degree of alignment ($\Theta_{average}$) of 47.5% ($\Theta_{average}$ = Br $_{forged}$ - Br $_{cast}$ / Br $_{cast}$)[9]. The intrinsic coercivity obtained here is higher then $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ sintered magnets (~20 KOe)[11] and considerably higher than that reported recently[10] of 18 KOe for hot pressed $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ magnets produced using an iron ring to constrain the specimen. Hot pressed magnets processed using a carbon steel ring achieved only around 14 KOe[12].

The variations of the magnetic properties of heat treated $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets as a function of strain rate, for a constant thickness reduction of 80%, are shown in fig. 2. There is a steadly rise in iHc as the strain rate is increased from 2.7X 10^{-2} to 3.0 X 10^{-1} s^{-1} at which the maximum value of iHc is reached. The highest value of iHc (~18 KOe) obtained is lowerthan the value of 21.5 KOe due to the lower thickness reduction used for the production of these magnets. The remanence and energy product follow a similar trend and saturate at a lower strain rate than the intrinsic coercivity. Therefore, it is considered empirically that the optimum strain rate is of the order of 2.0 X 10^{-1} s^{-1}. The remanence values and trends in behaviour observed in the present work are consistent with that previously reported for $Pr_{19}Fe_{74.5}B_5Cu_{1.5}$ hot pressed magnets[13]. The higher values of Br obtained with $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ hot pressed magnets[10] can be attributed to the lower strain rate and higher temperature used in that work. In general the densities obtained in the present work with the upset forged magnets were around 95% of the theoretical density.

Figure 3 shows the demagnetisation curves for an upset forged $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnet processed using a thickness reduction of 90% and at a strain rate of 8.0 X 10^{-2} s^{-1} as a function of post-forging annealing. In the as-upset forged condition (curve 1) quite reasonable magnetic properties are obtained and after annealing at 1000°C for 5 hours (curve 2) very little improvement was achieved. However, by further annealing at 500°C for 3 hours (curve 3) a significant increase in the iHc has been obtained. Small variations in the remanence have also been observed. This observations are consistent with a previous work[4] which has shown that most of the free iron, which exists in the cast alloy and is detrimental to the magnetic properties, is removed during the upset forged process (900°C). Thus, no significant increase in the iHc at high temperature annealing was observed in the magnets upset forged at 900°C. In the present work, however, a lower forging temperature (750°C) was used and it was expected that more free iron would be present after the upset forging process. This seems not to be the case however since only a small increase in iHc was achieved after the high temperature heat treatment and no low field shoulder was observed in the demagnetization curve. It was also shown[4] that lower temperature annealing (500°C) was beneficial to the magnetic properties and which also seems to be the case here for upset forged magnets processed at 750°C. The improvement with the lower temperature annealing (500°C) was attributed[4] to the grain boundary modification caused by this treatment which is just above the eutectic point of the grain boundary phase.

The microstruture of the as-upset forged $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnet is shown in Fig.4a, and the microstruture of this magnet after annealing at 1000°C for 5 hours is shown in fig.4b (demagnetization curves 1 and 2 in fig.3). The present sample was etched since the contrast between the different phases and grain boundaries was more pronunced in this condition. In the as-upset state, this magnet consists of matrix phase ($Pr_2Fe_{14}B$) and ill-defined grain boundaries. Free iron which existed inside of the matrix phase of the as-cast alloy has not been observed in this magnet. After annealing at 1000°C, the grain boundaries became slightly clearer, as seen in fig.4.b. A comparison between these two microstructure also demonstrates that there has been no significant changes during this heat treatment. This is in agreement with the magnetic properties measured for this magnet in both conditions (curves 1 and 2 in fig.3). The microstructure of this magnet after further annealing at 500°C is shown in the fig.4.c. Consistent with the intrinsic coercivity change, which increased substantially (curves 2 and 3 in fig.3), the microstruture also changed significantly. As can be seen clearly in this microstruture the grain boundaries became much more defined. This could indicate that the coverage of the matrix phase with a non ferro-magnetic Pr-rich material is improved after this low temperature heat treatment and this would enhance iHc since better isolation of the $Pr_2Fe_{14}B$ grains would be achieved. A comparison between these two microstructures also demonstrates, as expected, that there has been no significant grain growth during this low temperature heat treatment.

Figure 5 and 6 show the demagnetization curves for upset forged $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets processed at a strain rate of 8.0 X 10^{-2} s^{-1} using 88 and 84% of thickness reduction, respectively. In both cases there was a significant increase in coercivity with heat treatment (1000 + 500°C). The main difference between these magnets is found after the heat treatment (curves 2). At "88% of thickness reduction" a higher remanence is obtained whereas at "84% of thickness reduction" a higher intrinsic coercivity is achieved. This is consistent with previous work[2,13] which has shown that, as the thickness reduction is increased then the remanence of the magnets is also increased (probably because of a better c-axis grain alignment). It is well known for cast[6] and sintered[14,15] PrFeB magnets that improved grain alignment leads to a reduction of coercivity due to an increase of internal demagnetizing fields. This also seems to be the case for the upset forged magnets since the intrinsic coercivity is higher in the magnet processed with 84% of thickness reduction, in which poorer grain alignment is expected[13]. In the present magnets even using 90% of thickness reduction no high remanence has been obtained. Fig.7 shows that many grains are still misaligned in the magnet processed using 90% of thickness reduction, and this can probably be attributed to the low temperature used in the present work. Further studies are being carried out to optimize the pressing temperature and also to correlate the magnetic behaviour and microstruture of upset forged PrFeBCu magnets with sintered magnet produced by the hydrogen decrepitation (HD) process and based on the present composition[16].

Electron-probe microanalysis (EDX) in a scanning electron microscope has shown that the composition in the centre of the upset forged sample is similar to that close to the copper tube. This indicates that no marked preferential segregation of the liquid phase towards the edges has occurred during upset forging. A summary of the best magnetic properties obtained with the present magnets is given in Table 2. In this table has also been included the magnetic properties of forged magnets produced using the alloy in different starting conditions.

CONCLUSIONS

Anisotropic Pr-Fe-B-Cu-type permanent magnets have been produced from cast ingot materials using an upset forging process at a moderate forging temperature of 750°C. Magnets based on the composition $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ can achieve an intrinsic coercivity of higher than 21 KOe after post-upset heat treatments. The increase in the coercivity of $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets after a low temperature heat treatment can be attributed to the better magnetic isolation of the $Pr_2Fe_{14}B$ grains promoted with this treatment. The optimum strain rate determined for forging at a temperature of 750°C and at a thickness reduction of 80% was 2.0 X 10^{-1} s^{-1}. Two conditions were found for the thickness reduction: 84% for high coercivity and 88% for moderate remanence, at a constant strain rate of the 8.0 X 10^{-2} s^{-1}.

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ABSTRACT

Anisotropic Pr-Fe-B-Cu-type permanent magnets have been produced from cast ingot materials using upset forging. Magnets based on the composition $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ achieved an intrinsic coercivity higher than 21 KOe (>1674 KAm^{-1}) after post-forging heat treatments. A moderate forging temperature of 750 ^{0}C has been used in the present experiments. The upset forged magnets were annealed subsequently at 1000 ^{0}C for 5 hours and then at 500 ^{0}C for 3 hours. Strain rates (ε) from 2.7×10^{-2} to 3.0×10^{-1} s^{-1} and thickness reductions from 80 to 90 % have been employed. Losses of the praseodymium rich-phase during forging (squeezing out) were avoided by enclosing the specimens in a copper cylinder. An investigation of the magnetic properties of the cast ingots after the above annealing treatments has also been carried out for comparison.

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Table 1. Values of remanence, energy product and intrinsic coercivity for samples annealed at 1000^{0}C for 10 hours and then quenched (measured perpendicular to the growth direction).

Sample Number	Br (KG)	(BH) max (MGOe)	iHc (KOe)
1	5.67	6.30	6.28
2	6.20	7.70	7.66
3	5.65	5.94	6.10
4	5.29	5:59	7.03
5	5.47	6.03	7.40
6	5.58	6.10	6.44
Average	**5.64**	**6.28**	**6.81**

(Average error: Br:±0.05, BHmax:±0.05, iHc:±0.05)

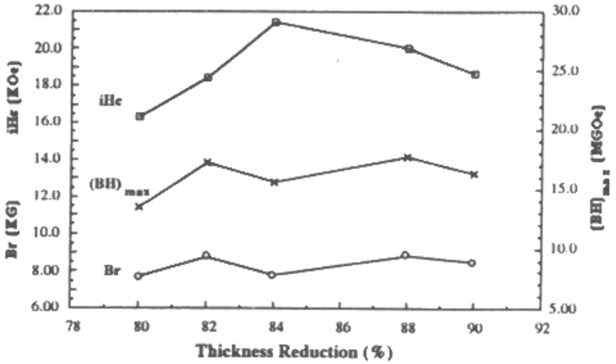

Fig.1 Variations of the magnetic properties of heat treated (1000 + 500^{0}C) $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged at 750^{0}C magnets as a functions of thickness reduction for a constant strain rate of 8.0×10^{-2} s^{-1}.

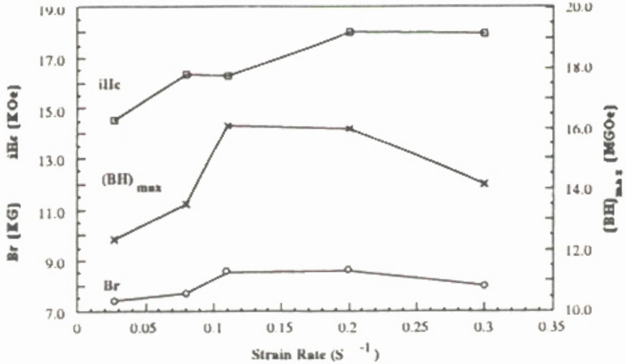

Fig.2 Variations of the magnetic properties of heat treated (1000 + 500^{0}C) Pr$_{20.5}$Fe$_{73.8}$B$_{3.7}$Cu$_2$ upset forged at 750^{0}C magnets as a functions of strain rate for a constant thickness reduction of 80%.

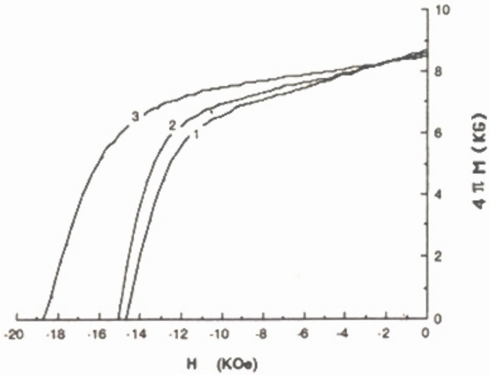

Fig.3 The demagnetization curves for upset forged at 750^{0}C Pr$_{20.5}$Fe$_{73.8}$B$_{3.7}$Cu$_2$ magnet using 90% of thickness reduction, as-upset forged (1), after heat treatment at 1000^{0}C (2) and after heat treatment at 1000^{0}C + 500^{0}C (3).

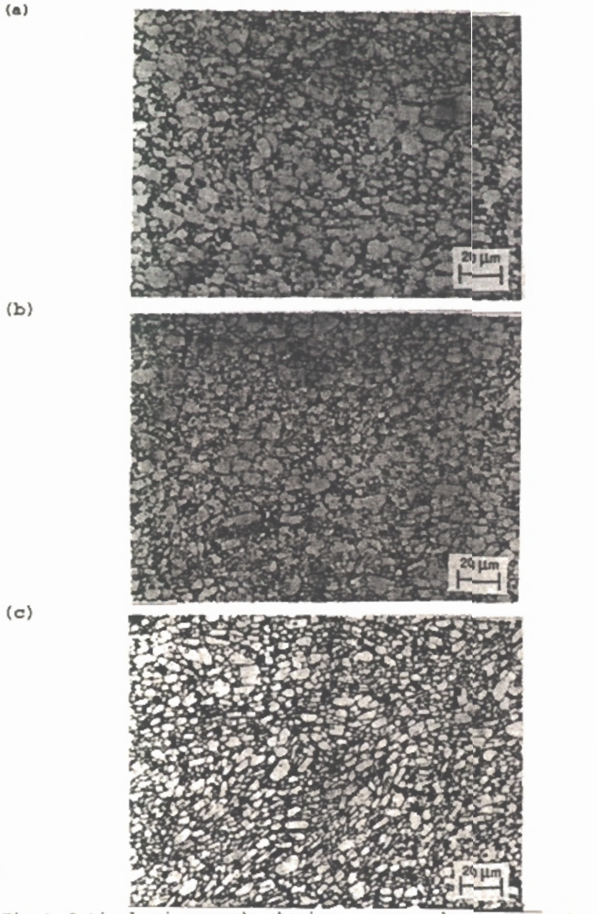

Fig.4 Optical micrograph showing a general view of the microstructure of the upset forged Pr$_{20.5}$Fe$_{73.8}$B$_{3.7}$Cu$_2$ magnet in the (a) as-forged, (b) heat treated at 1000^{0}C and (c) heat treated at 1000^{0}C + 500^{0}C condition (using nital 5%/1s).

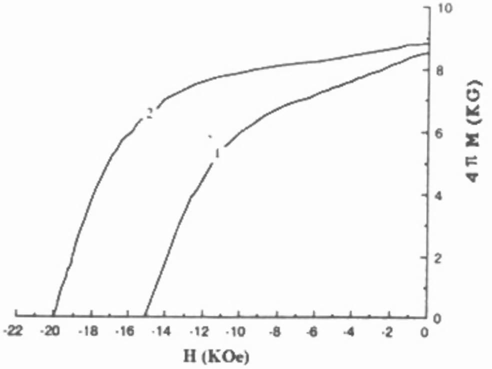

Fig.5 The demagnetization curves for upset forged (750°C) $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets using 88% of thickness reduction, before (1) and after heat treatment (2) for a constant strain rate of $8.0 \times 10^{-2} s^{-1}$.

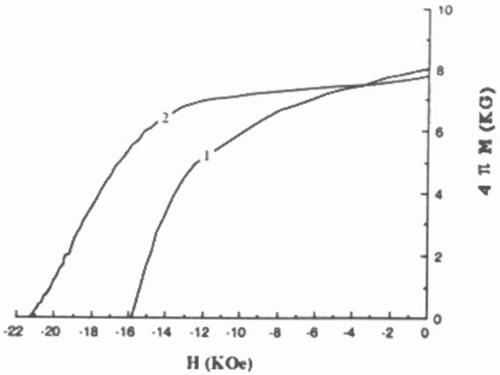

Fig.6 The demagnetization curves for upset forged (750°C) $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets using 84% of thickness reduction, before (1) and after heat treatment (2) for a constant strain rate of $8.0 \times 10^{-2} s^{-1}$.

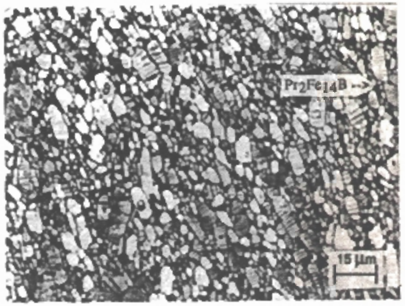

Fig.7 Optical micrograph showing a general view of the microstructure of the upset forged (750°C) $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnet in the heat treated at (1000°C + 500°C) condition (using ferrofluid).

Table 2. Values of remanence, energy product and intrinsic coercivity for forged magnets (750°C) using the $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ alloy in different starting conditions.

Start material	Processing conditions	Br [KG]	(BH) max [MGOe]	iHc [KOe]
Cast ingot	84% $8.0 \times 10^{-2} s^{-1}$	7.79	15.67	21.43
Cast ingot	88% $8.0 \times 10^{-2} s^{-1}$	8.85	17.81	20.04
HD-powder milled (on attritor for 5min)	75% $8.0 \times 10^{-2} s^{-1}$	6.54	9.90	14.50
HD-powder crushed (pestle and mortar)	75% $8.0 \times 10^{-2} s^{-1}$	7.23	12.07	16.37
HD-powder milled and aligned (on attritor for 5min)	70% $2.0 \times 10^{-1} s^{-1}$	5.50	6.89	11.36
Inside a die (cast ingot)	70% $8.0 \times 10^{-2} s^{-1}$	4.72	5.10	14.07
without copper ring (cast ingot	77% $2.0 \times 10^{-1} s^{-1}$	6.67	9.88	15.32