MAGNETIC PROPERTIES OF (Pr,Nd)-Fe-B-Cu MAGNETS PRODUCED BY UPSET FORGING OF CAST INGOT

P. V. P. Marcondes, A. N. Klein,
Laboratório de Materiais (LABMAT), EMC - UFSC ,
Florianopolis, S.C., Brasil.

R. N. Faria,
Instituto de Pesquisas Energéticas e Nucleares IPEN - CNEN,
CEP 05422-970 , São Paulo-SP

P. Bowen and I. R. Harris
School of Metallurgy and Materials, The University of Birmingham,
P.O.Box 363, Birmingham, B15 2TT United Kingdom

Anisotropic RE-Fe-B-Cu-type (RE:Pr,Nd) permanent magnets have been produced from cast ingot materials using upset forging. Magnets based on the composition $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ were produced with Br=10 KG, iHc=18.5 KOe and BH_{max}= 22.5 MGOe at a forging temperature as low as 790 ^{0}C; and after a post-forging heat treatment at 1000 ^{0}C for 5 hours and then at 500 ^{0}C for 3 hours. Hot pressing temperatures from 650 to 810 ^{0}C and a constant strain rate (ε) of 2.0 X 10^{-1} s^{-1} and thickness reduction of 85 % have been employed. Squeezing out of the RE-rich-phase during forging was avoided by enclosing the specimens in a copper ring.For comparison an investigation on the magnetic properties of $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets has also been carried out and a moderate energy product of 13.7MGOe has been achieved for this alloy.

INTRODUCTION

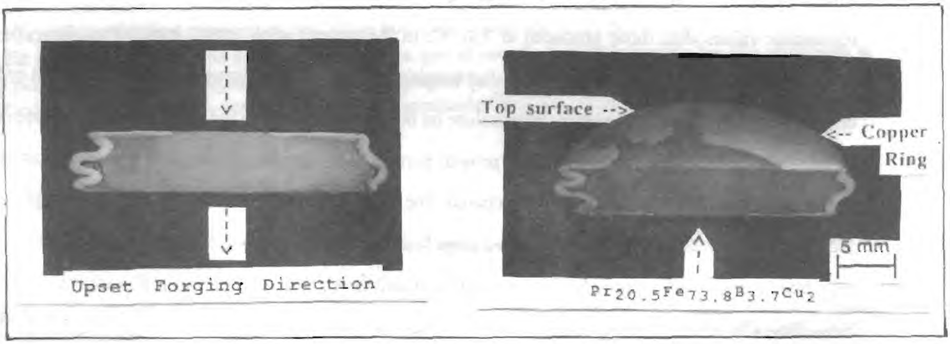
Recent work[1] has shown that magnets based on the composition $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ produced by upset forging at 750 ^{0}C can achieve a remarkably high coercivity of 21 KOe. In previous studies[2,3], upset forged $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets produced at a higher temperature of 900^{0}C showed slightly higher remanence values than those produced at 750 ^{0}C. In the present work, upset forged $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets have been produced and studied after forging at a range of temperatures (650-810 ^{0}C) in order to determine the influence of the upset temperature on the remanence. As in the previous work[1], a copper ring surrounding the cast ingot has been used to provide partial constraint and magnets of larger sizes, comparable to commercial scale magnets, have been prepared. The magnetic properties of the upset forged magnets have then been investigated before and after a two stage heat treatment. Using fixed upset forging parameters and heat treatments, magnets have also been produced and investigated using the following compositions: $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$, $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ (Seiko)[4,5] and $Pr_{16}Fe_{76}B_8$.

EXPERIMENTAL

The details of the preparation of the $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ cast alloys, the production of upset forged magnets and the upset forging equipment have all been described in previous papers[1-3,6-8]. Blocks (10X10 mm^2, height:40mm) cut from the ingot were embedded in a copper cylinder[1] and then upset forged at various temperatures under vacuum at a constant strain rate of 2.0×10^{-1} s^{-1} and an thickness reduction of 85 %. Rectangular magnets (8X8X5 mm^3) cut from the upset forged material were then pulsed in a magnetic field of 6 T and their second quadrant demagnetization curves determined. The magnets were then heat treated and their magnetic properties were then re-measured. The heat treatment of the forged ingot was carried out in vacuum at 1000 ^{0}C for 5 hours followed by quenching to room temperature, and then at 500 ^{0}C for 3 hours and again quenched to room temperature (600 ^{0}C for the Nd-based magnets). This heat treatment was a similar to that employed by Shimoda and co-workers[4,5].

RESULTS AND DISCUSSION

Figures 1a and 1b shows typical photographs of an ingot of the $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ alloy after upset forging. As can be seen, there are no visible cracks on the surface of the deformed material and it can also be seen that there was no rupture of the copper cylinder. For all the temperatures employed in the present work (650 to 810 ^{0}C), with a constant strain rate of 2.0×10^{-1} s^{-1} and thickness reduction of 85 %, no rupture of the copper cylinder has been observed. Cracks and bumps have been observed in Nd-Fe-B hot pressed magnets[9] when was not encapsulated by copper cylinder.



(a) (b)

Fig.1 Photograph of deformed ingot of a $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ alloy after the upset forging process.

The effects of forging temperature on the magnetic properties of the forged magnets, before and after heat treatment, are shown in figs. 2 and 3, respectively. The most striking feature of these graphs is the variation of the intrinsic coercivity with the forging temperature. Fig. 2 shows that there is a dramatic increase in iHc for the sample forged at 700^{0}C, which attained a value of 17.9 KOe without subsequent annealing. The remanence and energy product on the other hand exhibited similar behaviour with a regular change with forging temperature slowly increasing to a maximum at 790 ^{0}C. After the heat treatment at 1000 ^{0}C for 5 hours plus 500 ^{0}C for 3 hours, a small increase in the iHc (19.1 KOe) for the magnet forged at 700 ^{0}C was observed. The magnets forged at 700 ^{0}C all exhibited an appreciable increase in iHc after this heat treatment so that the sharp maximum at 700 ^{0}C was no longer appreciable. The dependence of Br and (BH)max on forging temperature appeared to be removed after the heat treatment. The higher (BH)max of 22.5 MGOe was achieved at 790 ^{0}C after the heat treatment.

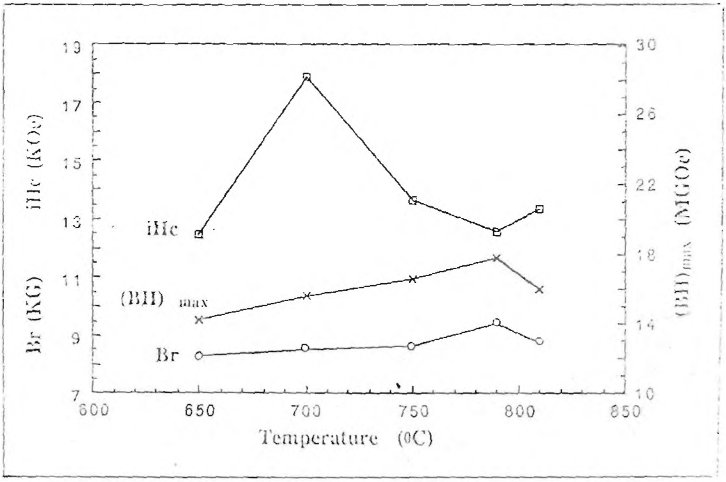


Fig.2 Variations of the magnetic properties of $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets as a function of forging temperature (without any heat treatment).

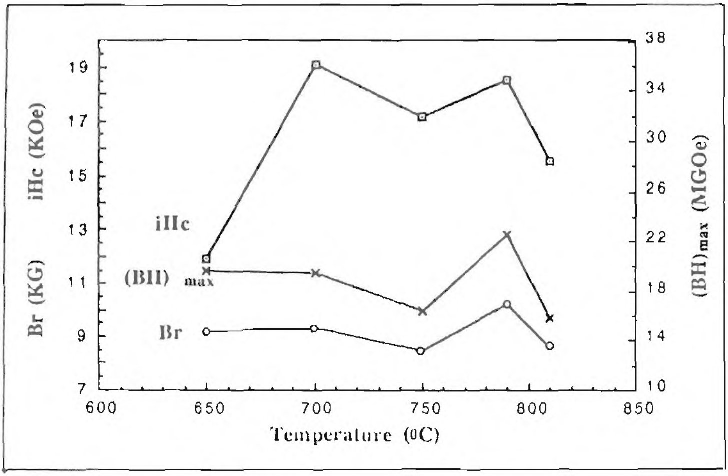


Fig. 3 Variation of the magnetic properties of heat treated $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets as a function of forging temperature (heat treatment : 1000 °C + 500 °C).

Figure 4 shows the demagnetization curves for magnets upset forged at 700 °C, before and after the heat treatment. The heat treatment proved to be effective not only in increasing the remanence and intrinsic coercivity but also in improving considerably the squareness factor (0.50).

The demagnetization curves for the magnets forged at 790 °C are shown in fig.5. In this case, a significant improvement in the intrinsic coercivity is noted with the heat treatment. The remanence reached 10.2 ± 0.05 KG for this magnet, which was the best value obtained in the present work. A very linear inductive coercivity for this magnet has also been observed. It has recently been reported[10,11], hot pressed magnets based on a $Pr_{19}Fe_{74.5}B_5Cu_{1.5}$ alloy, showed a remanence of 9.9 KG (in this case, no copper cylinder was used).

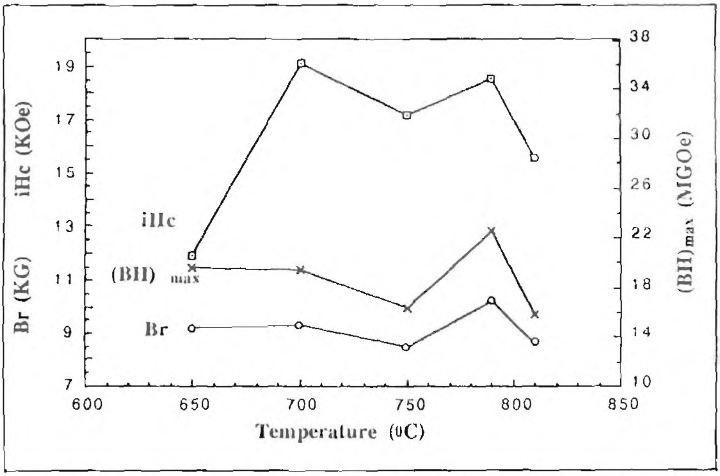


Fig.3 Variation of the magnetic properties of heat treated $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets as a function of forging temperature (heat treatment:1000^{0}C + 500^{0}C).

Figure 4 shows the demagnetization curves for magnets upset forged at 700 ^{0}C, before and after the heat treatment. The heat treatment proved to be effective not only in increasing the remanence and intrinsic coercivity but also in improving considerably the squareness factor (0.50).

The demagnetization curves for the magnets forged at 790 ^{0}C are shown in fig.5. In this case, a significant improvement in the intrinsic coercivity is noted with the heat treatment. The remanence reached 10.2 ± 0.05 KG for this magnet, which was the best value obtained in the present work. A very linear inductive coercivity for this magnet has also been observed. It has recently been reported[10,11], hot pressed magnets based on a $Pr_{19}Fe_{74.5}B_5Cu_{1.5}$ alloy, showed a remanence of 9.9 KG (in this case, no copper cylinder was used).

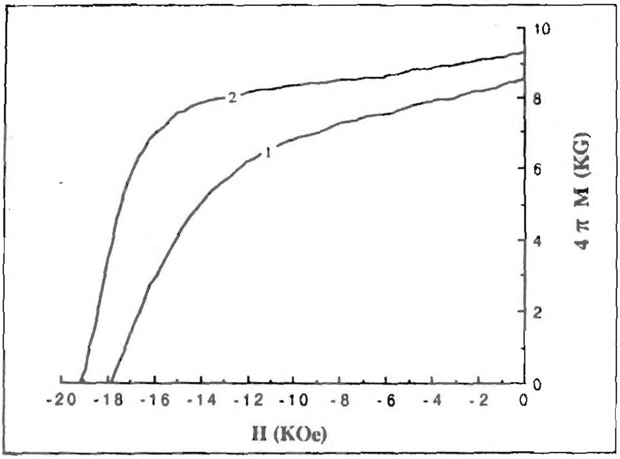


Fig.4 The demagnetization curves for upset forged (700°C) $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnet using 85% of thickness reduction and a strain rate of 2.0×10^{-1} s^{-1}, as-upset forged (1) and after heat treatment at 1000°C + 500°C (2).

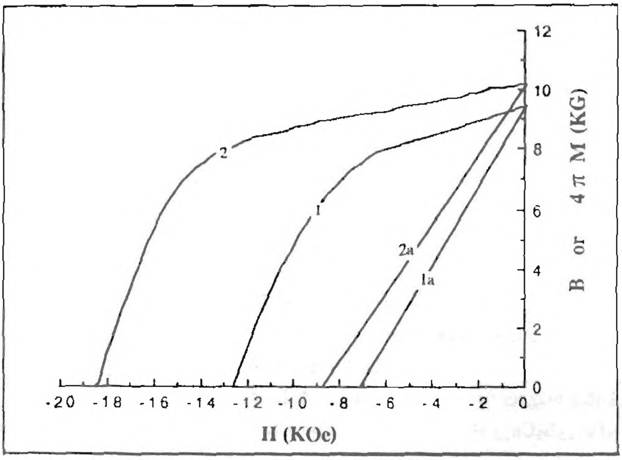


Fig.5 The demagnetization curves for upset forged (790°C) $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnet using 85% of thickness reduction and a strain rate of 2.0×10^{-1} s^{-1}, as-upset forged (1 and 1a) and after heat treatment at 1000°C + 500°C (2 and 2a).

The magnetic properties of a $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets processed using three different strain rates is shown in table 1. The best results were obtained with the highest strain rate used (2.0×10^{-1} s^{-1}). The demagnetization curves for this magnet, before and after the heat treatment, are shown in fig. 6. The main effect of the heat treatment was to improve the squareness factor. This behaviour is quite different from the $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets, which showed substantial increases (~ 27%) in coercivity with a similar heat treatment. The intrinsic coercivity and energy product of this $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnet are slightly superior to those reported by Shimoda at al[4,5] for an alloy with less rare-earth content ($Nd_{17}Fe_{76.5}B_5Cu_{1.5}$), although the remanence is slightly lower.

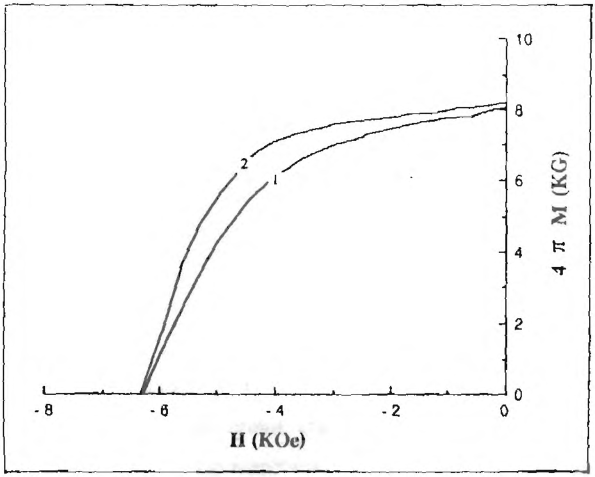


Fig.6 The demagnetization curves for upset forged (790°C) $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnet using 85% of thickness reduction and a strain rate of 2.0×10^{-1} s^{-1}, as-upset forged (1) and after heat treatment at 1000°C + 600°C (2).

Table 1. Values of remanence, energy product intrinsic coercivity and squareness factor for $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$ forged magnets (790°C) using a thickness reduction of the 85% and various strain rate (after heat treatment at 1000 °C+ 600 °C).

Strain rate	Br [KG]	(BH) max [MGOe]	iHc [KOe]	Squareness factor
2.7×10^{-2} s^{-1}	7.45	10.47	5.02	0.49
1.0×10^{-1} s^{-1}	7.44	10.58	5.23	0.49
2.0×10^{-1} s^{-1}	8.19	13.67	6.32	0.56

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

Table 2. Values of remanence, energy product, intrinsic coercivity and squareness factor for various types of forged magnets (790°C) using a strain rate of the 2.0×10^{-1} s^{-1} and a thickness reduction of the 85% (after heat treatment at 1000 °C + 500 °C).

Alloy composition	Br [KG]	(BH) max [MGOe]	iHc [KOe]	Squareness factor
$Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$	10.22	22.54	18.54	0.37
$Pr_{17}Fe_{76.5}B_5Cu_{1.5}$	7.79	11.06	6.00	0.40
$Pr_{16}Fe_{76}B_8$	8.18	10.53	6.53	0.24
$Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$	8.19	13.67	6.32	0.56

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

A summary and comparison of the best magnetic properties obtained in the present work for the various alloys is given in table 2. The upset forged magnets prepared using the $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ alloy showed undoubtedly the best magnetic properties. These magnetic properties are very similar to those obtained with sintered magnets based on this composition produced via hydrogen decrepitation (HD)[12,13]. In these HD magnets[12] the intrinsic coercivity also increased substantially after a similar heat treatment, reaching a iHc of

20 KOe. The $Pr_{17}Fe_{76.5}B_5Cu_{1.5}$ upset forged magnets showed poorer magnetic properties than the previous one, with higher Pr-content. This was a rather unexpected observation, since the magnetic properties reported for hot pressed magnets based on this composition (and similar processing conditions) by Shimoda at al[4,5,14,15] were much superior. However the present magnets were processed using a copper cylinder and in the previous work[4,5] the magnets were produced without a constraining device. Furthermore, according to this work[14] loss of the Pr-rich phase occurred in every sample, givening hot pressed magnets with a final composition of $Pr_{13.1}Fe_{80.4}B_6Cu_{0.5}$. In the present magnets the final analyzed composition did not change from the initial value ($Pr_{17}Fe_{76.5}B_5Cu_{1.5}$) and this indicates that different behaviour and consequently different magnetic properties should be expected. For instance the remanence enhancement associated with the loss of Pr will not be observed. The $Pr_{16}Fe_{76}B_8$ alloy showed very poor magnetic properties, confirming previous work[4], which showed that the magnetic properties deteriorate as the B-content is increased and the Cu-content is decreased. Table 2 also shows that a reasonable squareness factor was obtained with the $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets, similar to that of the $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ magnets but with a much lower value of iHc.

Further studies are being carried out to correlate the magnetic behaviour and the microstructure of upset forged PrFeBCu magnets with sintered magnet produced by the hydrogen decrepitation (HD) process and based on the present composition[16].

CONCLUSIONS

Anisotropic Pr-Fe-B-Cu-type permanent magnets have been produced from cast ingot materials using an upset forging process over a range of forging temperatures. The best magnetic properties were obtained for a forging temperature of 790°C, with a thickness reduction of 85% and a strain rate of $2.0 \times 10^{-1}\ s^{-1}$. Magnets based on the composition $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ exhibited a remanence > 10 KG after the post-upset heat treatments. The intrinsic coercivity of this $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnet increased substantially after a two stage heat treatment, reaching 18.5 KOe. The best energy product was 22.5 MGOe. $Nd_{20.5}Fe_{73.8}B_{3.7}Cu_2$ upset forged magnets showed a much smaller energy product of 13.7 MGOe.

ACKNOWLEDGMENTS

Many thanks are due to the CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico) and RHAE (Recursos Humanos em Areas Estratégicas) for the provision of a research grant (P. V. P. Marcondes). Thanks are due to REP (Rare Earth Products) for the provision of the alloys. Thanks are also due to the SERC, EURAM and CEAM (Concerted European Action on Magnets) for the support of the general research programme of which this work forms a part.

REFERENCES

[1] P. V. P. Marcondes, R. N. Faria, P. Bowen and I. R. Harris, to be published

[2] H. W. Kwon, P. Bowen and I. R. Harris, Proceedings of the 12th Int. Workshop on RE Magnets and their Applications Canberra, July 1992, 705

[3] H. W. Kwon, P. Bowen and I. R. Harris, J. Alloys and Compounds, 189 (1992) 131

[4] T. Shimoda, K. Akioka, O. Kobayashi and T Yamagami, J. Appl. Phys. 64 (10) (1988) 5290

[5] T. Shimoda, "Cast Pr-Fe-B Magnet" presented in the Global Business and Technical Outlook for NdFeB Magnet Markets organized by Gorham Advanced Materials Institute, California (USA), (1989), unpublished

[6] H. W. Kwon, P. Bowen and I. R. Harris, J. Appl. Phys. 70 (10) (1991) 6357

[7] R. N. Faria , J. S. Abell and I. R. Harris, J. Appl. Phys. 70 (10) (1991) 6104

[8] H. W. Kwon, P. Bowen and I. R. Harris, J. Alloys and Compounds, 182 (1992) 233

[9] K. Ohmori, L. Li and C. D. Graham, Jr., IEEE Trans. Magn. MAG 28 (5) (1992) 2139

[10] Z. Chen, Z. Chi, L. Wang and H. Fu., J. Appl. Phys. 71 (6) (1992) 2799

[11] Z. Chen, F. Xie,.Z. Chi, L. Wang and H. Fu., J. Appl. Phys. 70 (5) (1991) 2868

[12] R. N. Faria , J. S. Abell and I. R. Harris, J. Alloys and Compounds, 177 (1991) 311

[13] R. N. Faria , J. S. Abell and I. R. Harris, J. Alloys and Compounds, 185 (1992) 81

[14] T. Shimoda,.K. Akioka, O. Kobayashi, and T Yamagami, IEEE Trans. Magn. MAG 25 (5) (1989) 4099

[15] T. Shimoda,.K. Akioka, O. Kobayashi, and T Yamagami,, Paper No. 18P0101 at the 10th International Workshop on Rare-Earth Magnets and Their Applications, Kyoto, Japan, 16-19 May, 1989, p. 389

[16] P. V. P. Marcondes, R. N. Faria, P. Bowen and I. R. Harris, to be published