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The Effect of Zirconium Additions on the Magnetic Properties of Pr-Fe-B HD Sintered Magnets

R.N. Faria, H. Takiishi, L. F. C. P. Lima, I. Costa
Instituto de Pesquisas Energeticas e Nucleares,
IPEN-CNEN-S.Paulo, Brazil
M.R. Corfield, I.R. Harris
School of Metallurgy and Materials, University of Birmingham, Birmingham, B15 2TT, UK

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Abstract: Sintered permanent magnets based on the compositions $Pr_{16}Fe_{76}B_8$ and $Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ have been prepared using the hydrogen decrepitation process. It has been shown that small amount of zirconium addition to a $Pr_{16}Fe_{76}B_8$ sintered and fast cooled magnet improves the squareness factor (0.68 to 0.96) and diminishes the intrinsic coercivity. Sintered and slow cooled Pr-Fe-B-Zr and Zr-free magnets exhibited higher intrinsic coercivities (1095kA/m) than the fast cooled magnets.

Introduction

Recently, it has been reported that addition of zirconium to Pr-based permanent magnets produced via the hydrogenation disproportionation desorption and recombination (HDDR) process is an effective way of developing high anisotropy and good intrinsic coercivity in these magnets [1-4]. In the present work an investigation into the effect of zirconium addition and post-sintering cooling rates upon the magnetic properties of $Pr_{16}Fe_{76-x}B_8Zr_x$ (x=0 and 0.5) sintered permanent magnets has been carried out using the hydrogen decrepitation (HD) process.
In this work attempts have also been made to produce HD Pr-Fe-B-Zr sintered permanent magnets using a combination of two alloys with distinct compositions. This could also be a easier way of preparing laboratory sintered magnets with various compositions without the need of the lengthly casting process.

Material and Methods

HD sintered magnets were produced from as-cast $Pr_{16}Fe_{76}B_8$ and $Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ compositions using the isostatic pressing method. The alloys were powdered using a combination of the hydrogen decrepitation (HD) process [5,6] and milling in a roller mill for 20 hours. To produce a magnet with an average composition of $Pr_{14.5}Fe_{78}B_7Zr_{0.5}$ two alloys of compositions $Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ and $Pr_{13}Fe_{80.5}B_6Zr_{0.5}$ were decrepitated simultaneously and milled in the same manner. Subsequent HD powders were isostatically pressed and sintered either at 1060 or 1100°C for 1 hour under vacuum. Cooling from the sintering temperature was either achieved quickly, by placing a water-cooling jacket around the furnace vessel, or slowly, by leaving the vessel to cool in the furnace. Magnetic characterisation of the HD magnets was carried out using a permeameter, after full saturation in a field of 4.5 T.

Results and discussion

Fig. 1 and 2 show a comparison of the demagnetisations curves of fast and slow cooled HD PrFeB magnets, with and without zirconium addition (sintered at 1060°C). The most striking features of these curves are the diminution of the intrinsic coercivity with zirconium addition, in both cases, and the excelent loop shape of the zirconium containing fast cooled magnet. Both, $Pr_{16}Fe_{76}B_8$ and $Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ HD magnets show higher intrinsic coercivity with a slow cool treatment. Best remanence and squareness factor has been obtained with the fast cooled $Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ magnet.

Table 1 shows the magnetic properties of HD sintered magnets of $Pr_{16}Fe_{76-z}B_8Zr_z$ (z=0 and 0.5) compositions. The sintering treatments were either at 1060°C or 1100°C for 1 hour, followed by fast or slow cooling rates, depending on the particular sample. As observed in Fig. 1 and 2, zirconium has the effect of increasing significantly the squareness factor at the expense of a reduction in the intrinsic coercivity. Sintering at 1100°C had the same effect on the squareness factor without reducing the coercivity. Sintering the $Pr_{16}Fe_{76}B_8$ HD magnet at a higher temperature also improves considerably the density and remanence.

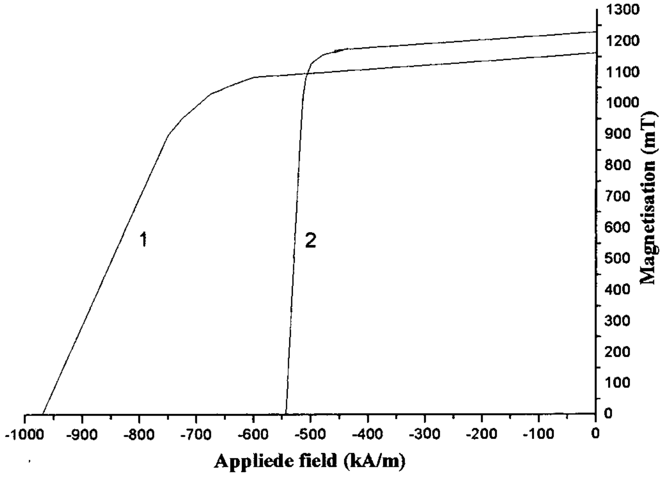

Fig. 1 – Permeameter demagnetisation curves for sintered (1) $Pr_{16}Fe_{76}B_8$ and (2)) $Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ magnets fast cooled from sintering.

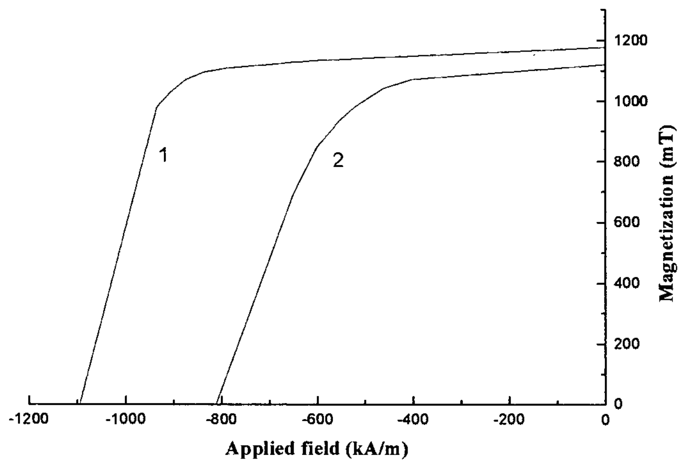

Fig. 2 - Permeameter demagnetisation curves for sintered (1) $Pr_{16}Fe_{76}B_8$ and (2) $Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ magnets slow cooled from sintering.

The intrinsic coercivity of HD Pr-Fe-B permanent magnets has been shown to be dependent on the cooling rate from the sintering temperature [7,8]. Slow cooling produces samples of higher intrinsic coercivity when compared to cooled samples relatively fast. Good squareness factor has been obtained for the $Pr_{14.5}Fe_{78}B_7Zr_{0.5}$ HD sintered magnet. Remanence is higher in this case, consistent with the reduced amount of praseodymium.

Table 1 – Magnetic properties of Pr-based HD magnets sinteres for 1 hour and fast or slow cooled. (error ± 2 %)

Alloys	Sintering Temperature [°C]	Cooling rate from Sintering temperature	Br [mT]	iHc [kA/m]	BHmax [kJ/m³]	SF [ratio]	dens. [g/cc]
$Pr_{16}Fe_{76}B_8$	1060	Fast	1162	967	253	0.68	7.34
$Pr_{16}Fe_{76}B_8$	1100	Fast	1217	968	278	0.82	7.39
$Pr_{16}Fe_{75.5}B_8Zr_{0.5}$	1060	Fast	1196	535	266	0.96	7.33
$Pr_{16}Fe_{76}B_8$	1060	Slow	1176	1095	254	0.80	7.35
$Pr_{16}Fe_{75.5}B_8Zr_{0.5}$	1060	Slow	1120	812	227	0.61	7.35
$Pr_{14.5}Fe_{78}B_7Zr_{0.5}$	1060	Fast	1229	573	277	0.90	7.35

Further work is being conducted to expand this study to Nd-Fe-B-Zr based magnets. In a first attempt a Zr added Nd-based ($Nd_{16}Fe_{75.9}B_8Zr_{0.1}$) HD sintered magnet prepared in our laboratory showed a rather low density (7.0 g/cc) and a squareness factor of only 0.89. Microstructural studies are underway in an attempt to explain the magnetic behaviour observed in this work for the Pr-based magnets.

Conclusion

In fast cooled samples zirconium has the effect of increasing the squareness factor of the HD Pr-Fe-B sintered magnets at the expense of the intrinsic coercivity. In contrast, slow cooled samples exhibited a decrease in magnetic properties with the addition of zirconium. It has been shown that is possible to produced a sintered magnet with good magnetic properties by a mixture of two alloys with distinct compositions.

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