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The Effect of Processing Parameters and Zirconium Additions upon the Magnetic Properties of Rare Earth Iron Boron HDDR Magnets

R. N. Faria , L. P. Barbosa , H. Takiishi,
I. Costa and L. F. C. P. Lima
Instituto de Pesquisas Energéticas e Nucleares,
IPEN-CNEN-S.Paulo, Brazil
luzinete@net.ipen.br

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Abstract: Hydrogenation disproportionation desorption and recombination (HDDR) magnets prepared from $RE_{16}Fe_{76-x}B_8Zr_x$ alloys (RE: Nd, Pr ; x=0-0.5) have been investigated in the present work. The effect of HDDR processing parameters, heat treatments, alloy composition and casting condition on the magnetic properties have been studied. It has been shown that the higher degree of anisotropy has been induced in the Nd-based alloy with 0.1 at% of zirconium. The optimum processing temperature for this alloy was close to 860°C.

Introduction

For many years isotropic and anisotropic HDDR powders have been studied and produced with Nd-Fe-B-type materials (see, for example [1-3]). In contrast, much less effort has been expended on Pr-based HDDR materials. However $Pr_{13}Fe_{81}B_6$ powders with lower magnetic properties (Br=670mT and iHc=549 kA/m) than Nd-based materials have been reported in the literature [4]. Recently, a $Pr_{13}Fe_{bal}Co_{24}B_6Ga_{1.0}Zr_{0.1}$ powder with good remanence but with rather a low coercivity has also been reported (Br=980 mT and iHc=382 kA/m) [5]. More recently, however, it has been shown that powders based on the composition $Pr_{13.7}Fe_{63.5}Co_{16.7}B_6M_{0.1}$ (M=Zr or Nb) could be produced with a high anisotropy and reasonable coercivity (Br=1032 mT and iHc=793 kA/m) after annealing the alloy and a relatively straightforward, subsequent HDDR treatment [6-9].
In the present work Nd and Pr-based magnets without Co and with a higher rare-earth content (16 at%) than previously studied have been investigated using various HDDR processing conditions and Zr additions.

Material and Methods

The commercial ingot alloys investigated in this work were prepared in a rectangular mould. To provide a comparison the alloys were studied in the as-cast state and also after a homogenization heat under vacuum at 1100°C for 20 h [5]. The HDDR treatment and details of the production of HDDR permanent magnets have all been described in previous papers [6-8]. Magnetic characterisation of the HDDR magnets was carried out using a permeameter. Measurements were performed after saturation in a pulsed field of 4.5 T. Remanence values have been normalised to assume 100% density (7.5 g/cc).

Results and Discussion

The effect of Zr additions

Fig. 1 shows the normalised remanence of HDDR $Nd_{16}Fe_{76-x}B_8Zr_x$ magnets processed at 800^oC and prepared using the isostatic pressing technique. An isotropic magnet has also been included for a comparison. Although in a small proportion, the best remanence is achieved when z is equal to 0.1 at.%. This result is consistent with previous studies [10], which have shown that, in cobalt containing HDDR magnets, the remanence and energy product of the bonded magnets have a similar behaviour. Table 1 shows a summary of the magnetic properties obtained with these magnets.

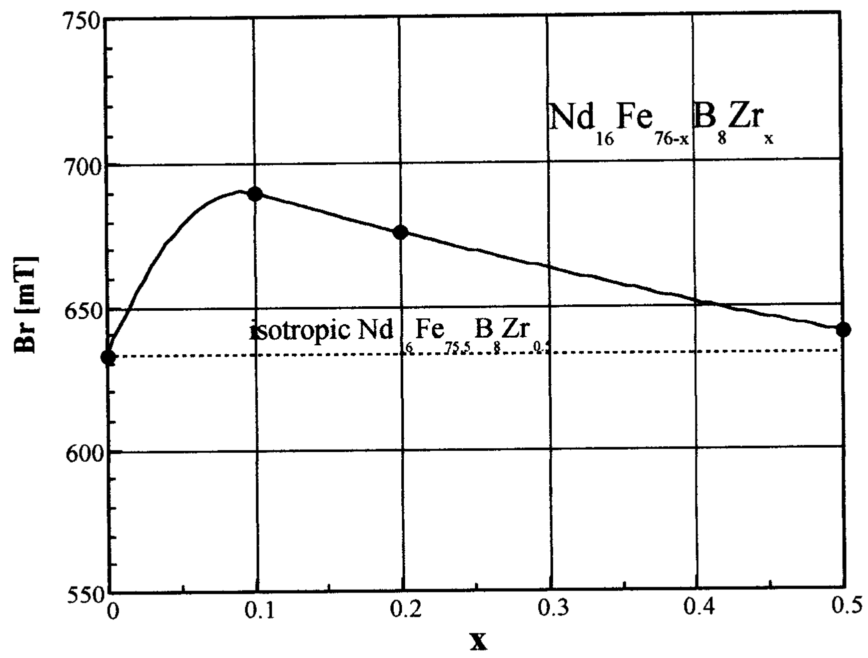

Fig. 1 – Normalised remanence of NdFeBZr magnets.

Table 1 - The effect of Zr additions on the magnetic properties of Nd-Fe-B HDDR magnets processed at 800°C.

STARTING ALLOY	Br [mT]	iHc [kA/m]	bHc [kA/m]	BHmax [kJ/m3]	Sq. [ratio]
$Nd_{16}Fe_{76}B_8$	633	791	387	66	0.30
$Nd_{16}Fe_{75.9}B_8Zr_{0.1}$	690	936	435	81	0.31
$Nd_{16}Fe_{75.8}B_8Zr_{0.2}$	676	869	421	76	0.31
$Nd_{16}Fe_{75.5}B_8Zr_{0.5}$	640	875	395	68	0.28
$Nd_{16}Fe_{75.5}B_8Zr_{0.5}$ isotropic	633	978	402	68	0.27

(measurement error: 2%)

Table 2 shows the magnetic properties of magnets processed at 800°C using three Pr-based alloys in the as-cast condition. A zirconium containing isotropic magnet has also been included for a comparison. Inferior magnetic properties have been obtained with these Pr-based magnets when compared to the Nd-counterpart shown in table 1.

Table 2 - Magnetic properties of Pr-based HDDR magnets.

STARTING MATERIAL (Processed at 800°C)	Br [mT]	iHc [kA/m]	bHc [kA/m]	BHmax [kJ/m3]	Sq. [ratio]
$Pr_{16}Fe_{75.5}B_8Zr_{0.5}$	617	622	335	59	0.30
$Pr_{16}Fe_{75.5}B_8Zr_{0.5}$ (isotropic)	554	621	308	48	0.28
$Pr_{13}Fe_{80.5}B_6Zr_{0.5}$	544	366	243	39	0.28
$Pr_{16}Fe_{76}B_8$	568	528	300	49	0.31

(measurement error: 2%)

The effect of casting conditions

Table 3 shows the magnetic properties of magnets processed at 800°C using a $Nd_{16}Fe_{75.5}B_8Zr_{0.5}$ alloy solidified using water cooled copper moulds with three distinct thicknesses. An isotropic magnet has also been included in the table for a comparison. The best remanence and intrinsic coercivity was obtained with the $Nd_{16}Fe_{75.5}B_8Zr_{0.5}$ alloy cast using a 12mm thick mould. The remanence in this case is comparable to that of the $Nd_{16}Fe_{75.9}B_8Zr_{0.1}$ magnet shown in Table 1.

Table 3 - The effect of ingot thickness (e) on the magnetic properties of $Nd_{16}Fe_{75.5}B_8Zr_{0.5}$ magnets processed at 800°C.

Ingot thickness (e)	Br [mT]	$_iH_c$ [kA/m]	$_bH_c$ [kA/m]	BHmax [kJ/m3]	Sq. [ratio]
15 mm	640	875	395	68	0.28
12 mm	680	876	417	77	0.30
5 mm	651	858	415	72	0.31
isotropic	633	978	402	68	0.27

(measurement error: 2%)

The effect of crushing the alloy ingot prior to processing

Table 4 shows the magnetic properties of HDDR magnets processed at 860°C using a $Nd_{16}Fe_{75.9}B_8Zr_{0.1}$ alloy in small lumps, after grounding the lumps to <500μm and also using the HDDR treatment twice (double HDDR). A decrease in remanence, energy product and squareness factor was observed in the latter cases, showing that the extra effort in crushing the alloy lumps or processing the material twice is unworthy for this particular alloy. Thus the grain refinement obtained during the HDDR treatment is not cumulative.

Table 4 - The effect of alloy state on the magnetic properties of $Nd_{16}Fe_{75.9}B_8Zr_{0.1}$ HDDR magnets.

STARTING MATERIAL	Br [mT]	iHc [kA/m]	bHc [kA/m]	BHmax [kJ/m3]	Sq. [ratio]
Cast alloy lumps	**756**	**1054**	**476**	**96**	**0.29**
Cast alloy grounded to < 500 μm	**634**	**1054**	**399**	**66**	**0.22**
HDDR material	**614**	**988**	**373**	**60**	**0.20**
Anisotropic sintered magnet	**637**	**715**	**377**	**66**	**0.30**

(measurement error: 2%)

In Table 4 the magnetic properties of a HDDR magnet prepared using as starting material a well orientated sintered magnet has also been included. Rather surprisingly this magnet showed the worst magnetic properties. It has been reported [11] that short time ball milling for HDDR $Nd_{14}Fe_{79}B_7$ powder, instead of the cast ingot, can result in moderate improvement in remanence and energy product, although a 5% reduction in coercivity is a non-avoidable effect of size reduction.

The effect of annealing the alloy

There has been an overall decrease in the magnetic properties of the HDDR magnets produced using alloys in the annealed condition for both, neodymium and praseodymium types. This behaviour is quite distinct to that reported [8] for cobalt containing magnets. This can be attributed to the higher amount of rare earth (RE) present in these magnets.

The effect of disproportionation temperature

To optimise the magnetic properties of the best composition found earlier a range of disproportionation temperatures has been investigated. Fig. 2 shows the normalised remanences and intrinsic coercivities of the $Nd_{16}Fe_{75.9}B_8Zr_{0.1}$ HDDR magnets as a function of the disproportionation (and recombination) temperatures. The optimum processing temperature was around 860°C, with the intrinsic coercivity peaking 1050 kA/m and the remanence reaching 756 mT. These results showed that the HDDR processing temperature is very effective in producing anisotropy.

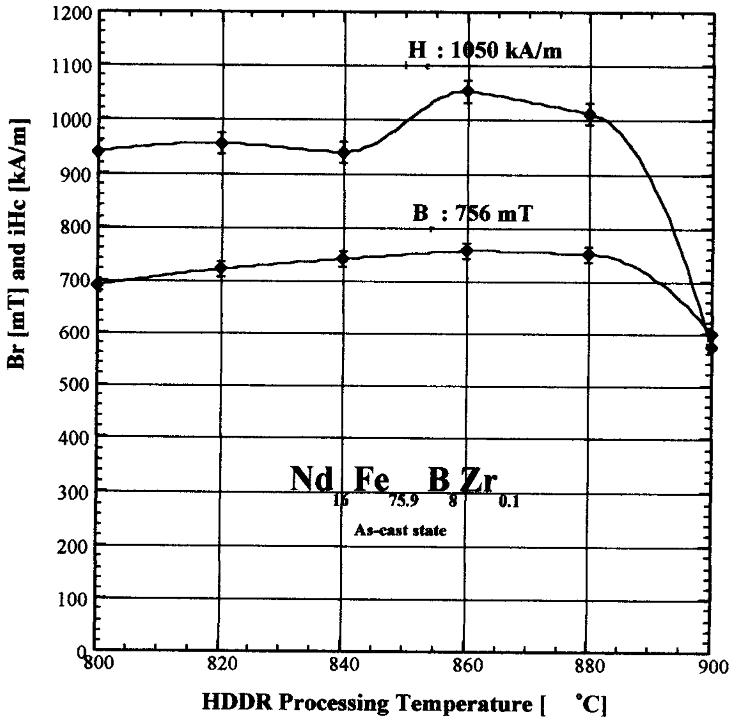

Fig. 2 – Normalised remanences and intrinsic coercivities vs processing temperature.

Conclusions

In the present study it has been shown that the as-cast alloy is the best starting material for HDDR magnets. Small aditions of Zr can induce a small degree of anisotropy in the Pr and Nd based magnets. Nd-based magnets exhibited superior mangnetic properties than their counterpart Pr-based magnets. The ideal temperature to process the $Nd_{16}Fe_{75.9}B_8Zr_{0.1}$ alloy was found around 860°C.

Acknowledgements

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