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Letter

Magnets produced by hot pressing $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe and $Nd(Fe,Mo)_{12}N_x$–α-Fe powders

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Abstract

High energy ball-milled and heat treated $Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$ (2:14:1 phase)–α-Fe and $Nd_{12}Fe_{75}Mo_{13}N_x$ (1:12 phase)–α-Fe magnetic powders have been compacted by hot pressing to form magnets. A systematic study on the effect of pressing temperature and time on the remanence J_r, intrinsic coercivity ${}_jH_c$, and density ρ, has been carried out. Hot pressing of $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe composite powder, containing a 40% volume fraction of magnetically soft α-Fe, produced an isotropic magnet with $J_r \approx 1$ T, ${}_jH_c \approx 428$ kA m^{-1} and $\rho = 7.59$ g cm^{-3}. The magnetic properties of near full density $Nd(Fe,Mo)_{12}N_x$–α-Fe-type magnets formed in a similar manner were also studied.

Keywords: Full density nanocomposite magnets; $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe; $Nd(Fe,Mo)_{12}N_x$–α-Fe; Hot pressing

1. Introduction

Remanence-enhanced composite materials, consisting of a hard magnetic rare-earth based phase and an α-Fe soft magnetic phase, are of considerable current interest because of their possible use as a permanent magnet. Hysteresis curves of isotropic nanocrystalline permanent magnet powders, numerically modeled by Kneller and Hawig [1], Schrefl et al. [2] and Skomski [3], show that the intrinsic coercivity ${}_jH_c$ and the enhanced remanent magnetic polarization J_r depend on the grain size of the soft magnetic phase. In order to achieve a significant enhancement of the J_r, and to preserve a high ${}_jH_c$, in isotropic nanocrystalline $Nd_2Fe_{14}B$-based magnets, a mean grain size of less than 20 nm is required [2]. Values of J_r in excess of 1.1 T and reduced remanent magnetic polarization, $\alpha = J_r/J_s$ (J_s is the saturation magnetisation) of 0.7, have been achieved in magnetic materials produced by high energy ball-milling (HEBM) and heat treatment, e.g. $Sm_2Fe_{17}N_x$–α-Fe [4], $(Nd,Tb)_2Fe_{14}B$–(FeCo,FeNb) [5] and $SmCo_7$–α-Fe [6].

Recent studies [7] have reported remanence enhancement in high energy ball-milled $Nd_{12.6}$-$Fe_{81.4-y-z}Co_yZr_zB_6$ (2:14:1 phase)–α-Fe ($y = 0$, 11.6; $z = 0$, 0.5) and $Nd_{12}Fe_{75}Mo_{13}N_x$ (1:12 phase)–α-Fe powders with a volume fraction of soft magnetic α-Fe phase of up to 75%. In the $Nd_2Fe_{14}B$–α-Fe powders, significant improvements in the intrinsic coercivity have been obtained with the addition of zirconium, due to reduction in the crystal grain size of the soft phase [7].

In this work we describe hot pressing techniques for the preparation of magnets from $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe and $Nd(Fe,Mo)_{12}N_x$–α-Fe powders, with a 40% volume fraction of magnetically soft α-Fe phase. A systematic investigation has been made on the effects of pressing temperature and time on the density, intrinsic coercivity and remanent magnetic polarization, in an attempt to obtain optimum magnetic properties.

2. Experimental

The $Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$ and $Nd_{12}Fe_{75}Mo_{13}$ samples were prepared by arc melting stoichiometric amounts of the constituent elements (99.9 at.% or better purity). Before HEBM the alloy lumps were crushed in an agate mortar to a particle size of less than 106 μm. The remanence-enhanced powders were prepared by HEBM the rare-earth-containing powder and Fe powder (purity 99.9 at.% from Aldrich Chemical Company Inc., particle size $\leq$10 μm) in a SPEX

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8000 Mixer Mill. The as-milled powders were heat treated in the temperature range of 600–850°C for 30 min under high purity argon to form either NdFeCoZrB (2:14:1 phase) or NdFeMo (1:12 phase) powders respectively. The crystallographic structures of the materials were determined by X-ray diffraction analysis using Cu Kα radiation. The nitrides were prepared by nitrogenation of $Nd(Fe,Mo)_{12}$–α-Fe powders at 450°C for 25 h under 100 kPa pressure of nitrogen.

The apparatus used for the hot pressing experiments has been described elsewhere [8]. Before each experiment both the pistons and the die were coated with boron nitride to prevent sticking of the sample. The apparatus was evacuated to 1.3 Pa, back filled with argon, and a precompaction pressure of 50 MPa was applied to the powder before inductively heating. During heating, the pressure was increased to 100–125 MPa and maintained for the duration of the process. Rapid cooling was achieved by blowing argon through the apparatus. In the series of experiments reported here, temperatures were varied over the range 700 to 860°C and pressing times from 5 to 60 s.

The magnetic properties were measured using a vibrating sample magnetometer (VSM) and/or a Quantum Design SQUID magnetometer. Experimental densities were determined by weighing under cyclohexane.

3. Results and discussion

Full details of the starting powders used in these hot pressing experiments, and their structure are contained in Ref. [7]. A summary of their magnetic properties are given in Table 1. Typically in the best powders, α is of order 0.70. Hysteresis loops for the starting $Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe powders, fixed in epoxy for the measurement, with a volume fraction of magnetically soft α-Fe phase of 50% and $Nd_{12}Fe_{75}Mo_{13}$–α-Fe epoxy bonded powders with a volume fraction of magnetically soft α-Fe phase of 40% are shown in Fig. 1.

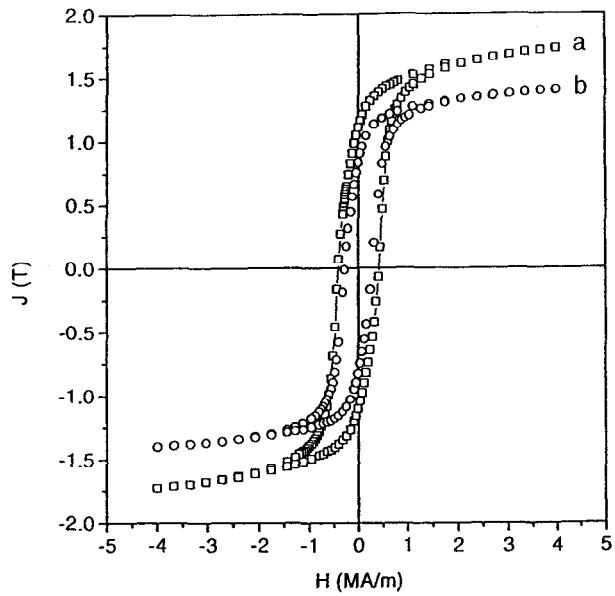

Fig. 1. Magnetic polarization J, as a function of the applied field H, for (a) $Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe expoxy bonded powders with a 50% volume fraction of magnetically soft α-Fe phase and (b) $Nd_{12}Fe_{75}Mo_{13}N_x$–α-Fe epoxy bonded powders with a 40% volume fraction of magnetically soft α-Fe phase (all data have been normalized to 100% density).

The variation in density of hot pressed $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe samples at a number of temperatures is shown in Fig. 2. Full densification (about 97 vol.%) in $Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe powders, with volume fraction of magnetically soft α-Fe phase of 40%, was achieved for $T \geq 830$°C. Fig. 3 shows the demagnetization curves for two $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe samples pressed at 830°C for 30 s and 860°C for 15 s. The lower pressing temperature resulted in a higher remanent magnetic polarization J_r. The samples processed at temperatures higher than 840°C and longer times show both a lower intrinsic coercivity ${}_jH_c$ and lower remanent magnetic polarization J_r, indicating partial decomposition of the starting material. Attempts at producing anisotropy by die-upsetting of $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe powders were unsuccessful. Plastic deformation was not observed in any of the samples within the temperature range 800–850°C. For mechanically alloyed NdFeB-type materials, the texturing of fully dense magnets by die-upsetting is only

Table 1
Saturation magnetic polarization J_s, remanent magnetic polarization J_r, reduced remanent magnetic polarization α, an intrinsic coercivity ${}_jH_c$, at room temperature of some HEBM $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe and $Nd(Fe,Mo)_{12}N_x$–α-Fe powders after optimal heat treatment.

Material	Volume fraction of magnetically soft α-Fe phase (%)	J_s (T)	J_r (T)	α (J_r/J_s)	${}_jH_c$ (kA m^{-1})
$Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe	0	0.96	0.67	0.70	680
$Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe	40	1.62	1.13	0.70	478
$Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe	50	1.73	1.11	0.64	440
$Nd_{12}Fe_{75}Mo_{13}N_x$–α-Fe	0	0.77	0.42	0.55	716
$Nd_{12}Fe_{75}Mo_{13}N_x$–α-Fe	40	1.37	0.85	0.62	281

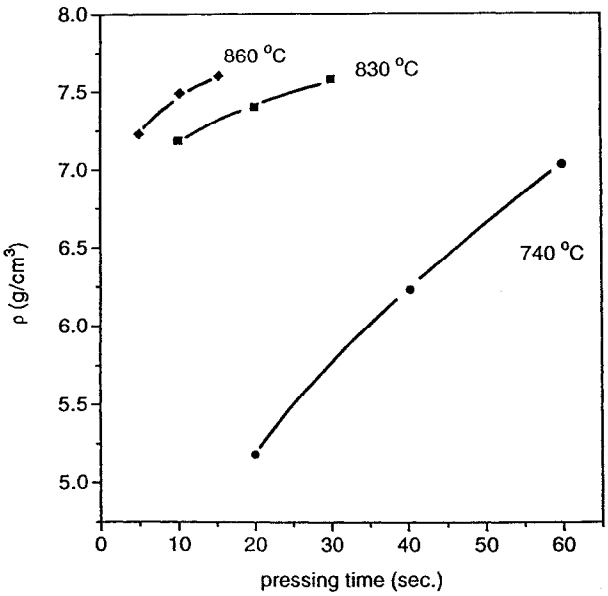

Fig. 2. The variation of density with pressing time for $Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe powders with a 40% volume fraction of magnetically soft α-Fe phase at a number of temperatures (lines are provided as guide to the eye).

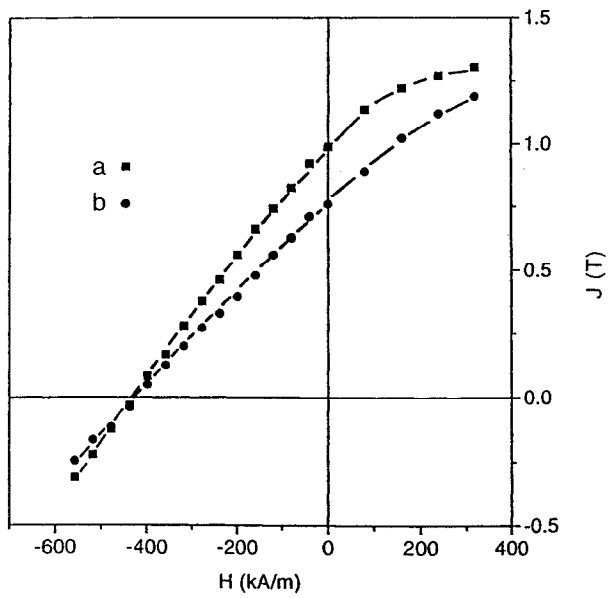

Fig. 3. Demagnetization curves of $Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe magnets with a 40% volume fraction of magnetically soft α-Fe phase pressed at: (a) 830°C for 30 s and (b) 860°C for 15 s.

possible with rare-earth concentrations of above, 14 at.% [9].

Magnets were also formed by hot pressing $Nd_{12}Fe_{75}Mo_{13}N_x$–α-Fe powders. Analysis of these magnets showed partial decomposition of the starting material to α-Fe and NdN during hot pressing and consequently a deterioration in the hard magnetic properties. A summary of the magnetic properties of hot pressed $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe and $Nd(Fe,Mo)_{12}N_x$–α-Fe magnets are given in Table 2.

Table 2
Experimental density ρ, remanent magnetic polarization J_r, and intrinsic coercivity $_JH_c$, at room temperature of some $Nd_2(Fe,Co,Zr)_{14}B$–α-Fe and $Nd(Fe,Mo)_{12}N_x$–α-Fe hot pressed magnets with a 40% volume fraction of soft magnetic α-Fe phase.

Material	Hot pressing parameters	ρ (g cm^{-3})	J_r (T)	$_JH_c$ (kA m^{-1})
$Nd_{12.6}Fe_{69.3}Co_{11.6}Zr_{0.5}B_6$–$\alpha$-Fe	740°C/60 s	7.03	1.05	441
	830°C/30 s	7.59	0.99	428
	860°C/15 s	7.65	0.76	419
$Nd_{12}Fe_{75}Mo_{13}N_x$–α-Fe	700°C/30 s	7.18	0.15	29

4. Conclusions

Two phase, isotropic magnets with $Nd_2(Fe,Co,Zr)_{14}B$ and α-Fe have been fabricated by hot pressing, with an intrinsic coercivity of 428 kA m^{-1}, remanent magnetic polarization of 1 T and density of 7.6 g cm^{-3}. Conversion to anisotropic material by hot die-upsetting was not achievable due to the low Nd content in the studied powders. Lower values of J_r and $_JH_c$ are obtained compared with epoxy bonded magnets produced from the same powders.

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