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Effect of the grain size on the magnetic properties of nanophase REFeB alloys

J.I. Betancourt R.[1], H.A. Davies*

Department of Engineering Materials, University of Sheffield, Sir Robert Hadfield Building, Mappin Street, Sheffield, S1 3JD, UK

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Abstract

The dependence of the remanence J_r and intrinsic coercivity ${}_iH_c$ on the grain size d_g for nanocrystalline RE (Nd–Pr)–Fe–B melt spun alloy ribbons was studied for single-phase stoichiometric 12% RE alloys and for nanocomposite Fe-rich 8% and 10% RE alloys. For single-phase alloys, ${}_iH_c$ decreases as the grain size diminishes, whereas for the two-phase nanocomposite alloys two different trends were observed: a rather constant dependency with decreasing d_g for 10% RE alloys (with 15% soft phase volume fraction) and an increasing trend for 8% RE alloys (with 30% soft phase volume fraction). The results are broadly in agreement with the predictions of micromagnetic modelling. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Remanence enhancement; Exchange spring magnets; Coercivity dependence

1. Introduction

The dependence of the magnetic properties (remanent polarisation J_r and intrinsic coercivity ${}_iH_c$) with grain size d_g has been reported previously for single-phase $Nd_{13.2}Fe_{79.6}B_6Si_{1.2}$ alloys [1]. In that report, J_r values of 0.8 T were found for $d_g \geqslant 40$ nm, consistent with the expected Stoner–Wohlfarth value for isotropic, uniaxial, randomly oriented non-interacting particles [2]. However, below this threshold d_g value, J_r increases progressively as d_g decreases, while ${}_iH_c$ is diminished. This effect results from the exchange coupling between the magnetic moments in neighbouring crystals, becoming more significant as d_g is reduced below the threshold value of ~40 nm, due to the enlargement of the interfacial area to volume ratio [3]. The threshold d_g is a function of the ratio A/K, where A is the exchange constant and K is the anisotropy constant. Micromagnetic calculations [4–7] showed good agreement with these experimental values, including the magnitude of the critical d_g. Further theoretical studies [8–11] predict that for nanocomposite alloys, which includes hard $RE_2Fe_{14}B$ grains and various volume fractions of soft α-Fe magnetic phase, both J_r and ${}_iH_c$ increase as d_g decreases below the critical d_g of ~40 nm for a given soft phase volume fraction. This paper provides experimental evidence that qualitatively confirms these predictions for nanocomposite RE(Nd–Pr)FeB/α-Fe alloy ribbons.

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*Corresponding author. Tel.: +44-0114-222-5518; fax: +44-0114-222-5943.

E-mail address: h.a.davies@sheffield.ac.uk (H.A. Davies).

[1] Present address: Instituto de Investigaciones en Materiales, UNAM, Apto Pos 70-360, Mexico D.F. 04510, Mexico.

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2. Experimental

The alloys in the compositional series $(Pr_{0.25}Nd_{0.75})_yFe_{94-y}B_6$ ($y = 8$, 10, 12 at%) were prepared by melting in an argon arc unit using pure component elements. The ribbon samples were produced by chill block melt spinning onto a copper roll, rotating at speeds within the range 16–25 m/s, in a high purity argon atmosphere within a sealed chamber. In each case, the alloy was rapidly melted in a quartz crucible using an r.f. coil and, at the appropriate temperature, ejected with an overpressure of 35 kPa of argon through a 0.6 mm diameter orifice onto the copper roll. The magnetic properties, the $J-H$ hysteresis loop, J_r, ${}_iH_c$ and the value of $(BH)_{max}$, computed from the derived BH loop, for ribbons having various thicknesses and various mean $RE_2Fe_{14}B$ crystallite diameters, were measured for each alloy with an Oxford vibrating sample magnetometer coupled to a 5 T magnet. Measurements were made in the plane of the ribbon samples and transverse to the spinning direction so that no correction for self-demagnetisation was necessary. The mean crystallite diameter was estimated by line broadening analysis using the Scherrer formula [12]. The microstructure of selected samples were investigated by transmission electron microscopy using a Philips 400 TEM, with thin foils prepared by ion-beam thinning.

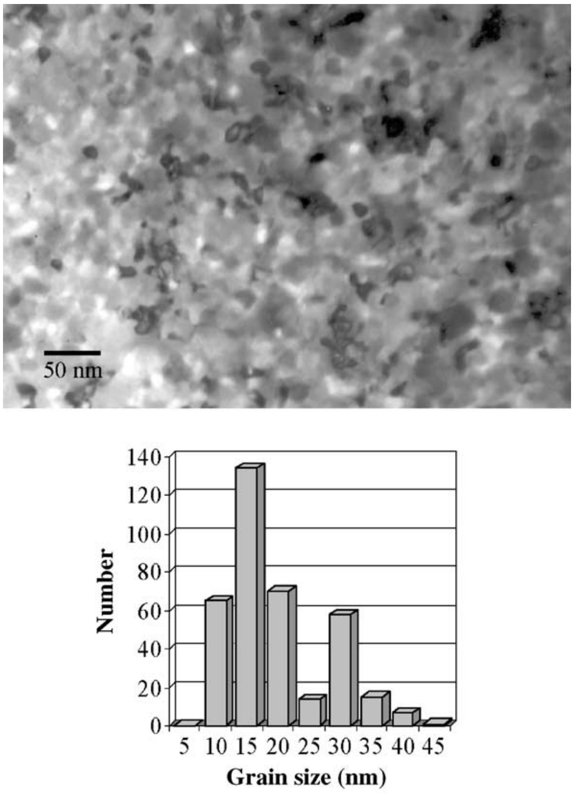

Fig. 1. Micrograph and grain size distribution of a selected sub-stoichiometric $(Nd_{0.75}Pr_{0.25})_8Fe_{86}B_6$ alloy, melt spun at 25 m/s.

3. Results and discussion

Transmission electron micrographs for selected samples, together with the grain size distributions, measured by the mean linear intercept method on three separate micrographs in each case, for the sub-stoichiometric $(Nd_{0.75}Pr_{0.25})_yFe_{94-y}B_6$ ($y = 8$, 10) alloy ribbons, rapidly quenched using roll speeds of 25 and 22 m/s, respectively, are displayed in Figs. 1 and 2.

For the 8% RE alloy sample (Fig. 1), a characteristic bimodal distribution is present. The peak centred around 15 nm corresponds to the α-Fe phase, which can be seen in the micrograph to be uniformly distributed throughout the microstructure. The second peak centred around 35 nm is ascribed to the hard $RE_2FE_{14}B$ grains. The soft phase volume fraction, calculated from the grain size distribution, is ~30%, in accord with previous reports for this composition in the $Nd_2Fe_{14}B$/α-Fe system [13,14]. For 10% RE alloy ribbon (Fig. 2), a bimodal grain size distribution is also evident, with peaks centred at 15 and 30 nm for the soft and hard grains, respectively. In this case, the soft phase volume fraction calculated from the grain size distribution is ~15%, in agreement with Mössbauer studies [13].

The dependence of J_r and ${}_iH_c$ on d_g is shown in Fig. 3. For the single-phase stoichiometric alloy ribbons, J_r increases from the Stoner–Wohlfarth value of 0.8 to 0.94 T as d_g decreases, due to the exchange coupling of the magnetic moments between adjacent grains. ${}_iH_c$, on the other hand, exhibits a corresponding reduction because of the

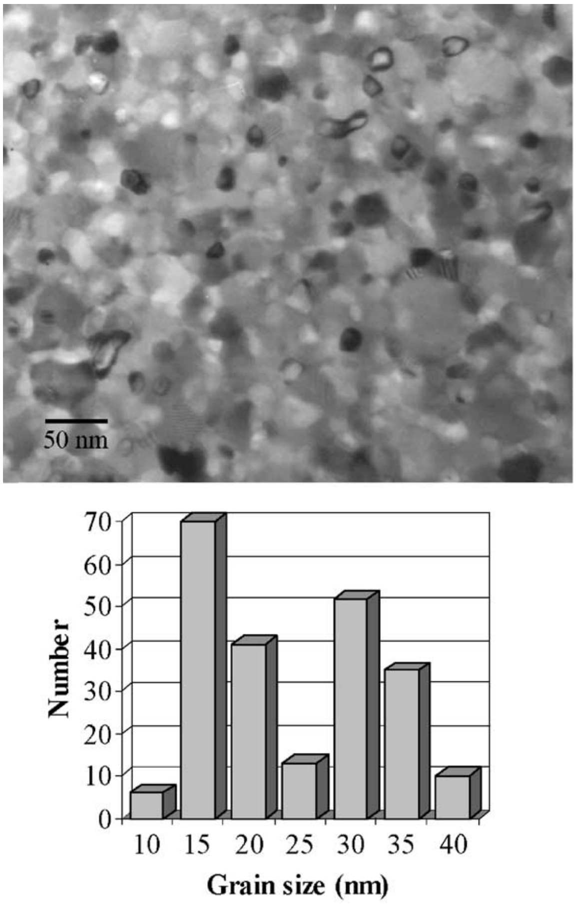

Fig. 2. Micrograph and grain size distribution of a selected sub-stoichiometric $(Nd_{0.75}Pr_{0.25})_{10}Fe_{84}B_6$ alloy, melt spun at 22 m/s.

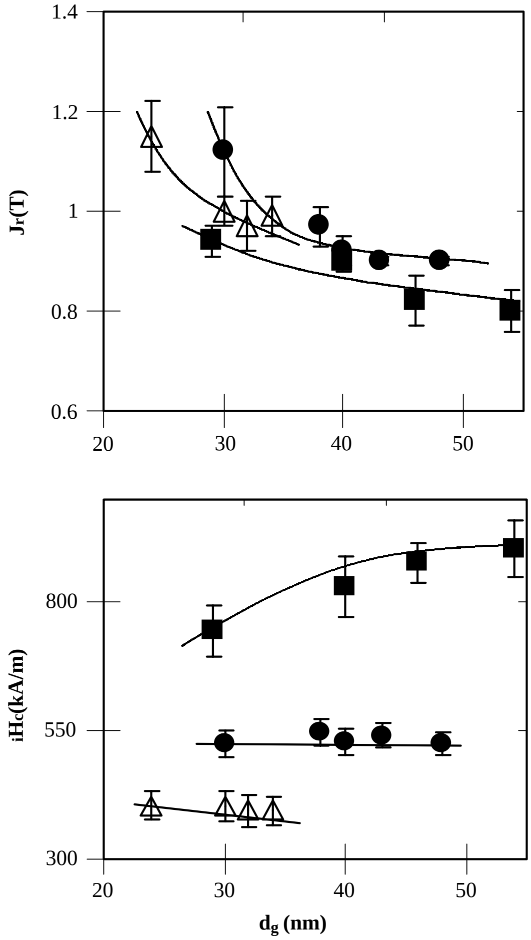

Fig. 3. J_r and ${}_iH_c$ dependence on d_g for $(Pr_{0.25}Nd_{0.75})_y$-$Fe_{94-y}B_6$, with $y=$ (i) 8% RE alloys (△), (ii) 10% RE alloys (●) and (iii) 12% RE (■) alloys.

reduction in the resistance to demagnetisation associated with the intergrain coupling.

For 10% RE alloy samples, J_r also exhibits enhanced values in excess of the corresponding non-interacting value (in this case 0.9 T), but ${}_iH_c$ shows a rather constant value around 520 kA/m, while for 8% RE alloy samples, an increasing trend of ${}_iH_c$ with diminishing d_g is evident, preserving an increasing J_r as d_g is reduced. Fischer et al. in discussing the results of their micromagnetic computations [8,9], stated that ${}_iH_c$ increases approximately logarithmically with decreasing d_g if the volume fraction of α-Fe is >50%. They added that, otherwise, ${}_iH_c$ first increases at small d_g. However, it is apparent from the present experimental data that the threshold volume fraction of α-Fe where a changeover occurs from decreasing ${}_iH_c$ to increasing ${}_iH_c$ with reduction in d_g is close to 15%. It is also apparent from the experimental data for 9.6% Nd (Billoni et al. [15]), 9.5% Nd (Zhang et al. [16]) and 10% Pr (Harland and Davies [17]) REFeB nanocomposite alloys that ${}_iH_c$ increases or at least does not decrease with diminishing d_g in cases where the volume fraction of α-Fe is below 20%.

The precise reason for the change in the dependence of ${}_iH_c$ on d_g at critical volume fractions of the soft phase is probably related to the effect of competition between the exchange-coupling characteristic of the hard and soft phases. However, J_r must also be influenced partly by the mean grain size for the soft phase which, experimentally, is known to be difficult to control independently of d_g for the hard phase.

4. Conclusion

An increasing coercivity with diminishing mean $RE_2Fe_{14}B$ grain size for REFeB/α-Fe nanocomposite melt spun alloys was observed experimentally for volume fractions of soft α-Fe phase greater than about 15%. This is qualitatively in accord with the predictions of micromagnetic modelling though the latter indicate that the threshold volume fraction would be much greater, of the order of 50%.

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