Microstructure and Magnetic Properties of $Nd_2Fe_{14}B/\alpha$-Fe Nanocomposite Prepared by HDDR Combined with Mechanical Milling

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Abstract

$Nd_2Fe_{14}B/\alpha$-Fe nanocomposite powders with a nominal composition of $Nd_{12}Fe_{82}B_6$ were prepared by HDDR combined with mechanical milling. The microstructure was studied by Mössbauer spectrometry and TEM. The magnetic properties were investigated by VSM using bonded magnet samples. The results showed that the annealing temperature had significant influence on both the recombination kinetics and the grain size of the $Nd_2Fe_{14}B$ and α-Fe phases, and the bonded magnets presented the best magnetic properties when the nanocomposite powders were prepared by annealing at 760 ℃ for 30 min.

Keywords: $Nd_2Fe_{14}B/\alpha$-Fe nanocomposite; HDDR; mechanical milling; microstructure and magnetic properties

1. Introduction

$Nd_2Fe_{14}B/\alpha$-Fe nanocomposites with exchange coupling between the hard and soft magnetic phases are attractive as promising permanent magnets [1-2]. These materials are usually prepared by melt spinning followed by an annealing treatment [3-4]. Unfortunately, the magnetic properties reported so far are much lower than expected [5-6].

HDDR is a well-established process to produce Nd-Fe-B alloy powders with sbmicron $Nd_2Fe_{14}B$ grains[7-9]. However, $Nd_2Fe_{14}B/\alpha$-Fe nanocomposites can not be produced by conventional HDDR. To develop alternative methods of producing nanocomposites, a new process which combines HDDR and mechanical milling has been proposed [10-11]. The present paper reports a study on the microstructure and magnetic properties of a $Nd_2Fe_{14}B/\alpha$-Fe nanocomposite by this new technique.

2. Experimental and results

The starting material was an as-cast $Nd_{12}Fe_{82}B_6$ alloy. The disproportionation was performed by mechanical milling for 20h in hydrogen with a pressure of above 0.2MPa using a QM-1SP planetary ball-mill. For a run of milling, 20g of alloy was milled. The ball to powder weight ratio was 20:1. The temperature for isothermal desorption -recombination annealing of the as-milled disproportionated alloy powders was 500 to 800℃, and the dwell time was 30min.

The phase constituents of the alloy powders were studied by transmission Mössbauer spectrometry with a ^{57}Co source. The microstructure and phase size were observed by TEM. The magnetic properties were measured by vibrating sample magnetometer (VSM) using Φ6×6mm bonded magnet samples.

Fig. 1 shows the Mössbauer spectra of the alloy powders subjected to milling and subsequent annealing at various temperatures. When the annealing was performed at 500℃, the spectrum was the same as that of the as-milled disproportionated powders, suggesting no desorption -recombination occurred (Fig.2(a)). After annealing at 650℃, the $Nd_2Fe_{14}B$ contribution was observed (Fig.2(b)). With the increase of the annealing temperature, the $Nd_2Fe_{14}B$ contribution increased drastically (Figs.2 (c) and (d)). Indeed, after vacuum annealing at 760℃, a complete recombination was achieved.

Fig. 2 shows representative TEM images of both the as-milled and the subsequently annealed alloy powders. The as-milled powders were fully disproportionated, with the average phase size being less than 8nm (Fig.3(a)). After annealing at 700℃, both newly formed $Nd_2Fe_{14}B$ grains with an average size of about 15-20nm and some retaining as-disproportionated phases were observed (Fig.3(b)). When the annealing was carried out at 760℃, the recombination was fully completed and the microstructure was featured by uniformly distributed $Nd_2Fe_{14}B$ and α-Fe grains of about 25nm in average size (Fig.3(c)). When the temperature was further raised to 800℃, the $Nd_2Fe_{14}B$ and α-Fe grains overgrew to about 40-50nm (Fig.3(d)).

Fig.3 shows the magnetic hysteresis loops of the bonded magnet samples prepared from $Nd_2Fe_{14}B/\alpha$-Fe nanocomposite powders obtained by annealing at various temperatures. Both the remanence and the coercivity increased with increasing annealing temperature up to 760℃, and then decreased. Since annealing at 760℃ for 30 min could achieve complete desorption-recombination while preventing the $Nd_2Fe_{14}B$ and α-Fe grains from overgrowth, the

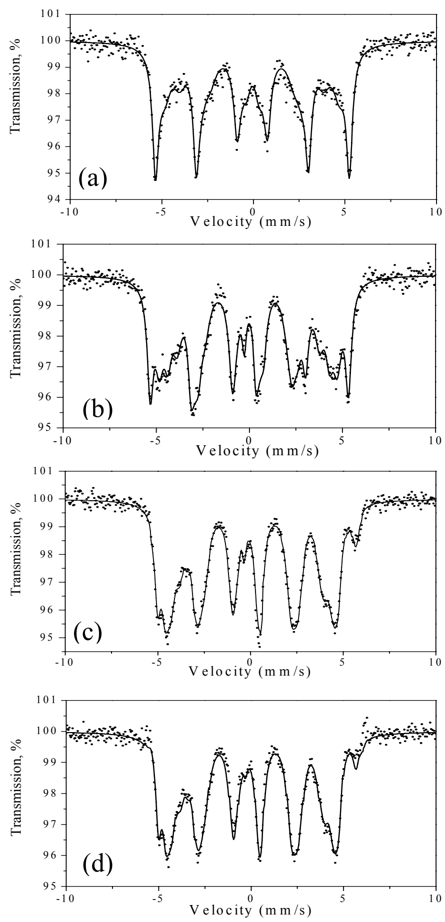

Fig. 1 Mössbauer spectra of alloy powders subjected to desorption-recombination at various temperatures: (a) 500℃; (b) 650℃; (c) 700℃; (d) 760℃

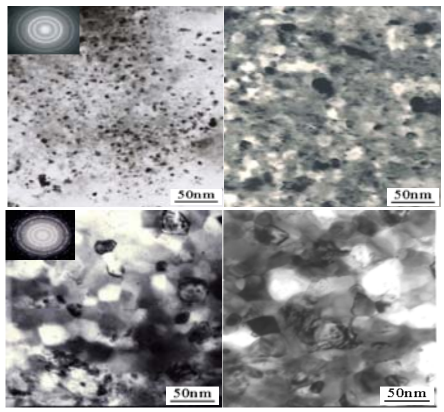

Fig. 2 TEM images of as-milled and desorption-recombination annealed alloy powders:

magnetic properties were correspondingly the best, with the remanence B_r, the coercivity ${}_bH_c$, and the energy product $(BH)_{max}$ achieving 0.72T, 465.8kA/m, and 90.6kJ/m^3 respectively.

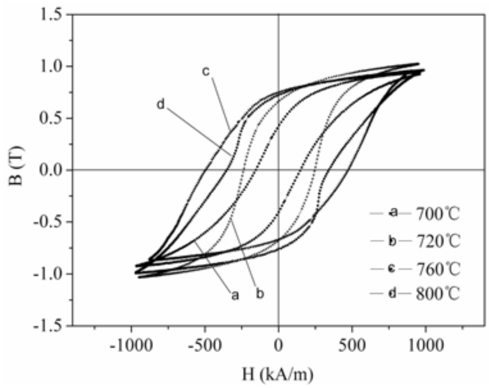

Fig. 3 Magnetic properties of bonded nanocomposite magnet samples

3. Conclusions

By mechanical milling in hydrogen for 20h, the $Nd_2Fe_{14}B$ phase in $Nd_{12}Fe_{82}B_6$ alloy was fully disproportionated into nano-structured Nd hydride, Fe_2B, and α-Fe with average size of less than 8nm.The recombination of the disproportionated phases occurred upon vacuum annealing at 650℃ with the formation of $Nd_2Fe_{14}B$ phase of 15-20nm. For a fixed processing time of 30 min, the optimal annealing temperature was 760℃, which gave rise to a fully recombined microstructure with $Nd_2Fe_{14}B$ and α-Fe phases of about 25nm. Correspondingly, the bonded magnet presented the best magnetic properties.

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