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Compositional optimization for nanocrystalline hard magnetic MRE–Fe–B–Zr alloys via modifying RE and B contents

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ABSTRACT

To reduce the rare earth content and maintain good magnetic properties for NdFeB based alloys, the effects of RE and B contents on the micro-structure and magnetic properties of nanocrystalline $MRE_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ ($MRE=Nd_{0.8}(Dy_{0.5}Y_{0.5})_{0.2}$, $y=0$–3) alloys have been investigated. Increasing B concentration leads to the appearance and increase of soft magnetic Fe_3B phase and reduced grain size. With decreasing MRE and increasing B concentrations, the coercivity decreased from 1159.8 kA/m for $y=0$ to 619.0 kA/m for $y=3$. The saturation magnetization and remanence increased with B content until $y=2$ then decreases. The B content also has effects on the exchange coupling, microstructure and thermal stability. While comparing $MRE_{10}Fe_{82.5}B_6Zr_{1.5}$ alloy with $MRE_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ ($y=1$ and 2) alloys, the alloy with 9 at% MRE can achieve similar magnetic properties as that with 10 at% MRE. The magnetic properties with coercivity of 792.2 kA/m, $(BH)_{max}$ of 128 kJ/m^3 and good thermal stability have been obtained for $MRE_9Fe_{79.5}B_{10}Zr_{1.5}$ alloy.

1. Introduction

The current research on NdFeB based permanent magnets mainly focused on both optimization and compositional modification process. Compared to sintered NdFeB alloys with rare earth (RE) rich composition, the nanocomposite NdFeB has relatively low RE contents and thus, relatively low cost, which is preferable for industry. The NdFeB based nanocomposite alloys include two types of $Nd_2Fe_{14}B$/α-Fe and $Nd_2Fe_{14}B/Fe_3B$ alloys based on the type of soft magnetic phase. Except the low cost, they have also shown good properties with high remanence J_r, high maximum energy product $(BH)_{max}$, and good temperature stability. Hence, these alloys are important compositions for bonded and die-upset magnets. The $Nd_2Fe_{14}B$/α-Fe nanocomposite has been extensively investigated [1], and generally, reducing RE content enhances the remanence but degrades the coercivity if the optimal nanostructure can be realized. As for $Nd_2Fe_{14}B/Fe_3B$ type nanocomposite alloys, a good balance between the J_r and $_jH_C$ can be obtained in the composition range of $Nd_{10-7}Fe_{bal.}B_{10-15}$ [2–4]. Betancourt et al. [5] obtained high magnetic properties in melt spun $(Nd_{0.75}Pr_{0.25})_{10}(Fe_{0.9}Co_{0.1})_{78}Nb_2B_{10}$ alloy with $_jH_C=1042$ kA/m, $J_r=0.88$ T, and $(BH)_{max}=123$ kJ/m^3. Chiu et al. [6] obtained the best properties of $J_r=0.95$ T, $_jH_C=859.4$ kA/m, and $(BH)_{max}=142$ kJ/m^3 in $Pr_9Fe_{77.5}Ti_{2.5}B_{11}$ alloy. Using 10 at.% Co substituting Fe, not only the Curie temperature T_C of both 2:14:1 phase and α-Fe phase can be increased, the temperature stability and magnetic properties are also improved. Its magnetic properties are $J_r=0.97$ T, $_jH_C=899.2$ kA/m and $(BH)_{max}=147$ kJ/m^3. In addition, with 10.5 at% B content in $(Nd_{0.95}La_{0.05})_{9.5}Fe_{68}Co_{10}Cr_2B_{10.5}$ alloy, high combined magnetic properties with $J_r=1.04$ T, $_jH_c=756$ kA/m, and $(BH)_{max}=158$ kJ/m^3 were obtained [7]. In all these alloys, addition of B was found important to the microstructure improvement [6].

Also, with the intention to increase properties/price ratio, we previously report the optimized nanocrystalline $MRE_{10}Fe_{86}B_6$ with mixed rare earth ($MRE=Nd_{0.8}(Dy_{0.5}Y_{0.5})_{0.2}$) alloy with excellent room temperature magnetic properties and thermal stability [8]. In this composition, 50% Dy was substituted by Y, which not only reduces the content of high cost element Dy, but also maintains good magnetic properties and enhances the temperature stability. In addition we found that 1.5% Zr doping employed in this alloy can not only suppress the precipitation of the unwanted 2:23:3 phase [9], but also can enhance the coercivity. Based on this previous work, here we study the effects of RE and B contents on the magnetic properties of this alloy. The aim of this work is to optimize the $MRE_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ alloys by modifying B and MRE contents and to achieve both good magnetic properties and low materials cost.

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2. Experimental

Nanocrystalline $MRE_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ (MRE=$Nd_{0.8}(Dy_{0.5}Y_{0.5})_{0.2}$, $y=0$, 1, 2, and 3) alloys were prepared by argon arc melting followed by melt spinning. Raw material ingots of Nd, Dy, Y, Fe, Fe–B and Zr with purities higher than 99.8% were arc melted to produce small buttons under Ar atmosphere. The buttons were turned and re-melted six times to ensure homogeneity. All examined as-spun ribbons were produced at an optimized wheel speeds varied between 12 and 18 m/s depending on the composition. For $MRE_7Fe_{79.5}B_{12}Zr_{1.5}$ alloys, the melt spinning wheel speed varied between 2 and 18 m/s due to its high glass forming ability. The phase constitution was characterized by X-ray diffraction (XRD, Philip *X-pert*) using Cu-Kα radiation. The microstructures of the ribbons were characterized by the transmission electron microscopy (TEM, JEOL JEM-2100 transmission electron). The magnetic properties of directly quenched alloys were tested by physical property measurement system (PPMS, Quantum Design Co., USA) equipped with a vibrating sample magnetometer (VSM) using a maximum magnetic field of 8 T. For all measurements, the ribbon was placed with the applied field in the ribbon plane and across its width. Since the thickness of the ribbon (20–40 μm) is much smaller than the width and length (~mm), the direction of the measurement has a demagnetization factor close to zero. Therefore, No demagnetization factor is considered in this work. Magnetization–temperature (*M*–*T*) curve was measured at a magnetic field $H=1000$ Oe between 300–1000 K.

3. Results and discussion

3.1. Effects of RE and B concentrations on the phase structure and microstructure

The X-ray diffraction patterns for $[Nd_{0.8}(Dy_{0.5}Y_{0.5})_{0.2}]_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ alloys are shown in Fig. 1. For $y=0$, all peaks come from 2:14:1 phase and no other phase was detected due to the relatively high MRE content, although with tiny amounts of both α-Fe and Fe_3B phases may exist in this alloy. With the increase of B content, the MRE content decreases, the soft magnetic Fe_3B can be well detected, as indicated for $y=3$. The grain sizes estimated from XRD patterns are shown in Table 1. The results indicate that increase of B is beneficial for grain refinement, which has advantage for improving magnetic properties of the nanocrystalline alloys. The selected TEM images for the alloys with $y=1$ and $y=2$ are

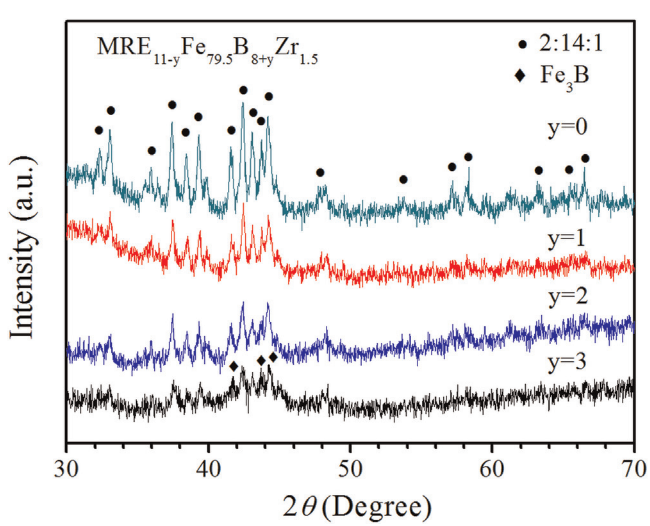

Fig. 1. XRD patterns of the $MRE_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ alloys with $y=0$, 1, 2, and 3.

Table 1
The grain size, room temperature magnetic properties and estimated phase constitution for $MRE_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ alloys.

y	Grain size (nm)	J_r (T)	H_{ci} (kA/m)	$(BH)_{max}$ (kJ/m^3)	Phase constitution, vol%	
					$MRE_2Fe_{14}B$	Fe_3B
0	~50	0.80	1159.8	108	97.3	2.5
1	~66	0.86	864.4	124	90.3	9.5
2	~39	0.89	792.2	128	83.1	16.9
3	~37	0.87	619.0	107	75.5	22.0

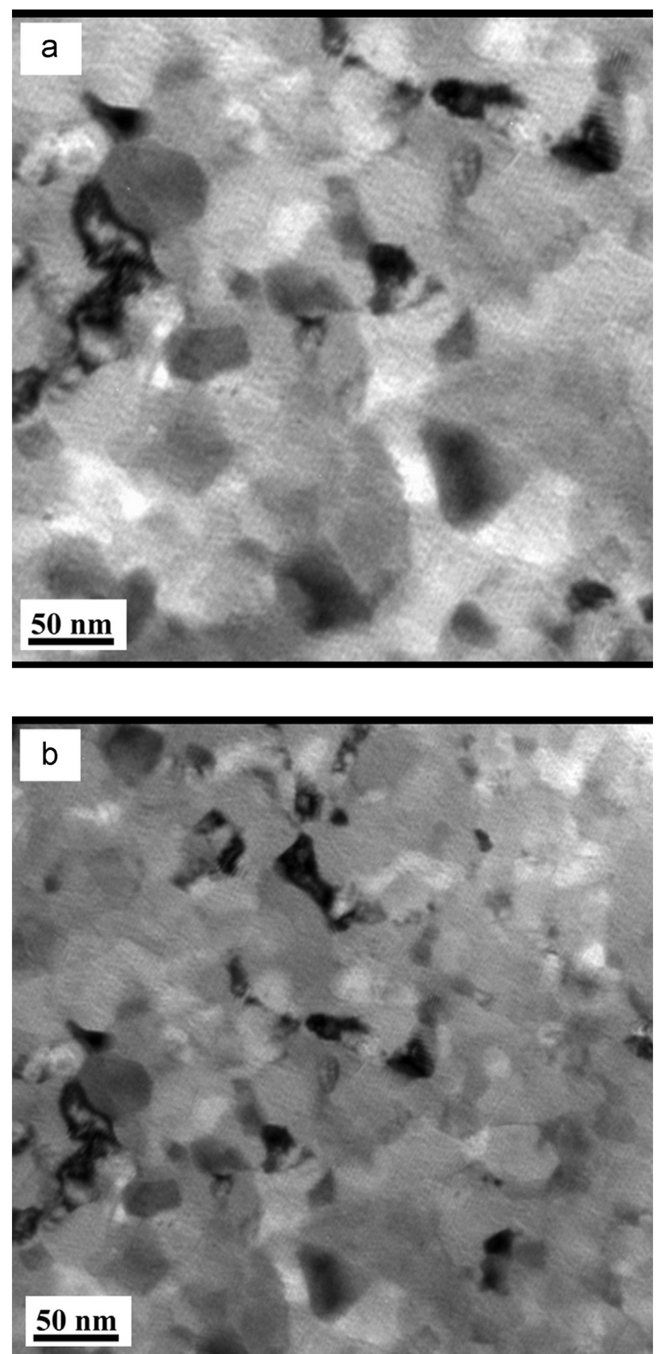

Fig. 2. TEM images for the $MRE_{11-y}Fe_{79.5}B_{8+y}Zr_{1.5}$ alloys with $y=1$ (a) and $y=2$ (b).