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Short communication

Study of microstructure and correlative magnetic property in bulk $Fe_{61}Nd_{10}B_{25}Nb_4$ permanent magnet

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ABSTRACT

The correlation between microstructure and magnetic property of a bulk $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy are investigated. The microstructure of the as-cast $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy shows a small amount of NbFeB phase with a grain size of 500 nm embedded in an amorphous matrix. The as-cast sample shows soft magnetic behavior at room temperature, after a heat treatment the hard magnetic properties are observed. A fully dense bulk $Fe_{61}Nd_{10}B_{25}Nb_4$ permanent magnet is obtained with an intrinsic coercivity ($_iH_c$) of 1191 kA/m and a maximum energy product ($(BH)_{max}$) of 31.7 kJ/m^3 after annealing at 943 K for 20 min. The corresponding microstructure consists of $Nd_2Fe_{14}B$, $NdFe_4B_4$ and NbFeB phases. The existence of the hard magnetic $Nd_2Fe_{14}B$ phase is the reason resulting in a high value of $_iH_c$. On the other hand, the influences of $NdFe_4B_4$ and NbFeB phases in the annealed specimen on the magnetic properties are also discussed.

1. Introduction

In 2002, Zhang et al. reported that a fully dense $Nd_2Fe_{14}B$/(Fe_3B, α-Fe) nanocomposite permanent magnet was produced by crystallization of a bulk glassy $Fe_{67}Co_{9.4}Nd_{3.1}Dy_{0.5}B_{20}$ alloy with a diameter of 0.5 mm [1]. The hard magnetic behavior of this alloy was attributed to ferromagnetic exchange coupling between nanoscale soft and hard magnetic phases. Compared with a traditional way to prepare Nd–Fe–B permanent magnets including melt spinning or mechanical milling process, the fully dense magnets have obvious advantages, such as low cost, casting in precise dimensions easily and avoiding the polymer dilution of magnetic phase. Hence, Zhang's work [1] paves a promising way for the bulk permanent magnet prepared by a simple process of copper mold casting and subsequent heat treatment, which stimulates further investigations of microstructure and magnetic properties in other bulk Fe-based alloy systems [2–13]. For example, the fully dense bulk (Fe,Co)–Zr–(Pr,Dy)–B or (Fe,Co)–Zr–(Nd,Y)–B permanent magnets with the shapes of rods, sheets or tubes were obtained by a devitrification annealing process [5–7,9–11]. However, the glass forming ability (GFA) of these alloys is not good enough to get bulk amorphous alloys with a large size, e.g. only 1 mm diameter of rod was obtained [5–7]. Moreover, the value of $_iH_c$ of these alloys is not very impressive, less than 400 kA/m, because of the coarser microstructure or the existence of non-ferromagnetic phases. In order to improve the GFA and magnetic properties, process optimization and the compositional modification have been employed. Recent studies [12,13] found that bulk (Pr,Dy)–(Fe,Co)–B–Zr–Ti and Nd–Fe–Nb–B hard magnets with the coercivity of more than 750 kA/m could be fabricated by a rapid quench method. Especially, a quaternary fully dense $Fe_{64.32}B_{22.08}Nd_{9.6}Nb_4$ hard magnet with the coercivity approaching as high as 1100 kA/m was developed [8]. However, an increase in the coercivity is usually accompanied with a sacrifice of remanence (B_r), resulting in a low energy product.

It is well known that the coercivity and remanence are extrinsic magnetic properties and they are intimately related to the microstructure of a magnetic material. Thus a number of structural investigations of fully dense bulk Fe-based permanent magnets are carried out to explore the influences of the microstructure on the magnetic properties [4,8,10,12,13]. So far, there are two opinions elucidating the relationship between the microstructure and coercivity in these materials. On one hand, a uniformly distributed hard magnetic $Nd_2Fe_{14}B$ phase or Pr_2(Fe, Co)$_{14}$B phase was believed to be responsible for high value of the coercivity (>770 kA/m) [8,12,13]. On the other hand, fine grain sizes and ideal volume fractions of soft and hard magnetic phases presented in the Nd–Y–Fe–Nb–B magnet was reported to the reason for the optimal hard magnetic properties, such as $_iH_c$ = 891.52 kA/m, B_r = 0.57 T and $(BH)_{max}$ = 56.8 kJ/m^3 [4]. However, the magnetic properties of fully dense bulk Fe-based permanent magnets are not impressive because the alloy having an optimal microstructure has not been developed so far. Therefore, it needs to further investigate the relationship between the microstructure and the hard magnetic behavior of fully dense Fe-based magnets.

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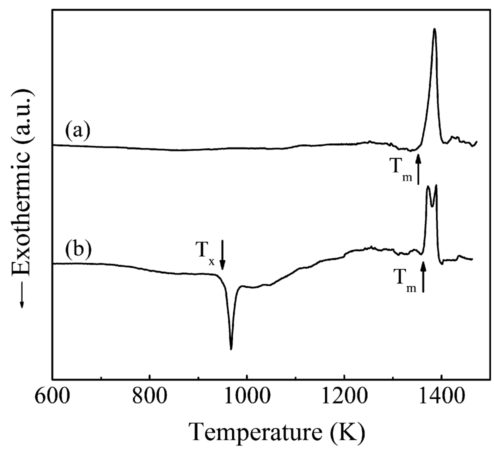

Fig. 1. DSC curves of as-cast $Fe_{65}Nd_{10}B_{25}$ alloy (a) and as-cast $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy (b).

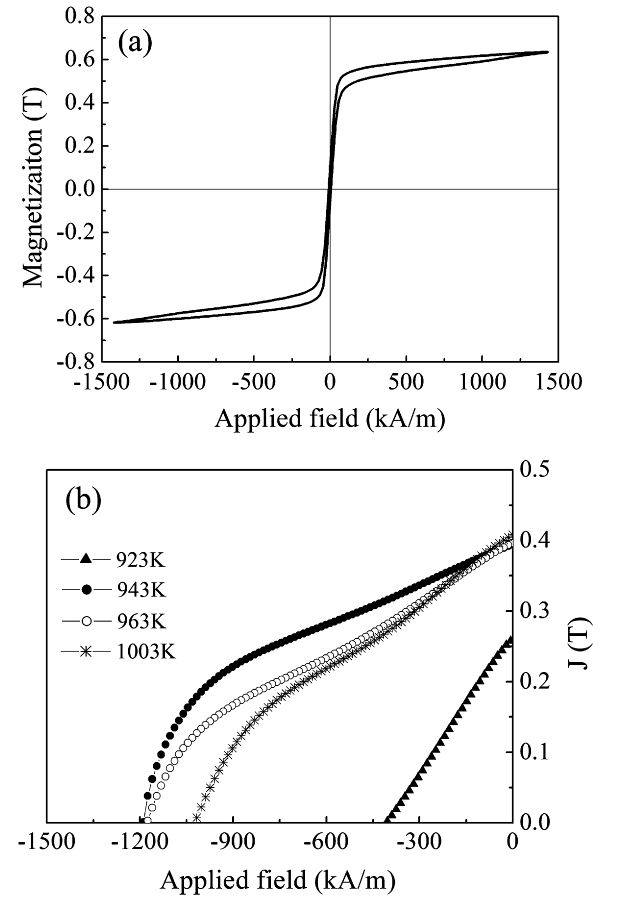

Fig. 2. Hysteresis loop of the as-cast $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy (a), and demagnetization curves of the samples annealed at 923 K, 943 K, 963 K and 1003 K for 20 min, respectively (b).

In this work, we investigate the relationship between the microstructure and magnetic properties of the $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy. On the basis of the ternary Nd–Fe–B alloy, Nb is chosen because it can enhance GFA significantly [8,14]. On the other hand, refining and homogeneous microstructure can be formed because of Nb enrichment at grain boundaries or possibly the formation of Nb–Fe–B intergranular phase between magnetic phases which give rise to enhanced magnetic properties [15–19]. This work would be helpful to improve magnetic properties and develop future applications of fully dense bulk Fe-based permanent magnets. The role of Nb element is also discussed.

2. Experimental procedures

The ingots with compositions of $Fe_{65}Nd_{10}B_{25}$ and $Fe_{61}Nd_{10}B_{25}Nb_4$ (at.%) were prepared by arc melting the mixture of pure metals Fe, Nd, Nb and Fe–B alloy in an argon atmosphere. Bulk sheet specimens (10 mm in width, 80 mm in length and 1 mm in thickness) were prepared by suction casting of the molten ingots into a water-cooled copper mold. The samples were annealed at 923 K, 943 K, 963 K and 1003 K for 20 min in a vacuum furnace with a gas pressure of less than 2×10^{-3} Pa. The thermal parameters were determined by a differential scanning calorimetry (DSC) using a NETZSCH DSC 404 calorimeter under a heating rate of 20 K/min. Structural investigations were carried out by X-ray diffraction (XRD) using a D/max-2550 diffractometer with Cu Kα radiation and by a JEM-2010F transmission electron microscopy (TEM). The Curie temperature (T_C) of the magnetic phase was determined by thermogravimetric analysis (TGA) using a Pyris l TGA under a heating rate of 10 K/min. Magnetic properties were measured by a vibrating sample magnetometer (VSM) with a maximum applied field of 1.8 T at room temperature. The density of the $Fe_{61}Nd_{10}B_{25}Nb_4$ sample is 7.305 g/cm^3 using Archimedes principle. Specimens for the magnetic measurements are 4 mm in width, 4 mm in length and 1 mm thickness, being cut from the sheet sample. The demagnetizing factor is 0.135. Before measuring demagnetization curves, all the annealed samples were saturated with a 4 T pulsed magnetic field.

3. Results

The DSC trace for the as-cast ternary $Fe_{65}Nd_{10}B_{25}$ alloy does not show any indication for an exothermic signal [see Fig. 1(a)], which suggests no amorphous phase formed. At the temperature of 1353 K, an endothermic signal corresponds to melting (marked as T_m) is observed. For the as-cast $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy, a single sharp exothermic peak is clearly observed [see Fig. 1(b)], suggesting that a certain amount of amorphous phase is contained in this alloy. The onset temperature of crystallization, T_x, is 955 K. The sharp endothermic peaks in the range of 1300–1400 K are resulted from a multi-step melting behavior. The onset temperature of melting is also marked as T_m.

To further investigate the transition from soft magnetic behavior to hard magnetic behavior, annealing treatment of the as-cast $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy is carried out. Based on the DSC result (see Fig. 1), four annealing temperatures of 923 K, 943 K, 963 K and 1003 K are selected. The hysteresis loop for the as-cast $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy and demagnetization curves for the annealed samples are shown in Fig. 2(a) and (b), respectively. The as-cast sample presents soft magnetic behavior, while all annealed samples show hard magnetic properties. It is seen in Fig. 2(b) that the coercivity and the squareness of demagnetization curves increase significantly when the annealing temperature is below 943 K. With further increasing annealing temperature, the coercivity and the squareness of demagnetization curves decrease. The $Fe_{61}Nd_{10}B_{25}Nb_4$ alloy annealed at 943 K for 20 min exhibits the optimum magnetic properties, i.e. ${}_iH_c$ = 1191 kA/m, B_r = 0.42 T and $(BH)_{max}$ = 31.7 kJ/m^3.

The XRD patterns of the as-cast and annealed $Fe_{61}Nd_{10}B_{25}Nb_4$ alloys are shown in Fig. 3. For comparison, the XRD pattern of the ternary $Fe_{65}Nd_{10}B_{25}$ alloy is also included, which shows a coexistence of the $Nd_{1.1}Fe_4B_4$ phase, α-Fe phase and $Nd_2Fe_{14}B$ phase [see Fig. 3(a)]. No significant amorphous phase is found in the $Fe_{65}Nd_{10}B_{25}$ alloy, which is in coincide with the DSC trace [see Fig. 1(a)]. For the as-cast $Fe_{61}Nd_{10}B_{25}Nb_4$ sample [see Fig. 3(b)],