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Effects of aging at room temperature on as-spun $Nd_2Fe_{14}B/\alpha$-Fe nanocomposite magnets

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Abstract

As-spun exchange-coupled Nd–Fe–B–X (X = In, Ti) nanocomposites were prepared by using the melt-spinning method. Effects of aging at room temperature on magnetic behaviors of the as-spun powders were investigated. It was found that both coercivity H_c and reduced remanence m_r are significantly enhanced as a result of short-range atom diffusion and stress relaxation at the intergranular region, rather than of grain growth or crystallization of amorphous phase, after being stored for a long time at room temperature (0–35 °C). Enhancements reach a maximum when amorphous and crystalline phases coexist in nanocomposites. XRD results indicate that a transitional $Nd_2Fe_{14}B$-like intergranular phase may be formed or developed near grain boundaries during the aging process. It is a more crystallographically coherent, stress relaxed, and chemically balanced intergranular region that is very beneficial to the promotion of intergrain exchange coupling and hardening between adjacent magnetic soft and hard grains. The role of the intergranular phase in magnetic exchange coupling and hardening mechanisms is also discussed in this paper.

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1. Introduction

Exchange-coupled $Nd_2Fe_{14}B/\alpha$-Fe nanocomposites have been widely explored due to their peculiar properties and potential application prospects. It is essential to obtain a fine, uniform phase distribution and adequate intergranular structure in order to achieve good magnetic performance for this kind of nanocomposite magnet. Optimal microstructures are usually composed of a well exchange-coupled magnetic soft phase with higher magnetization such as α-Fe,

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and a magnetic hard phase with higher anisotropy, such as $Nd_2Fe_{14}B$ [1]. A unique feature of this nanocomposite magnet is the presence of an appropriate intergranular amorphous phase when optimum magnetic properties are obtained. Across the intergranular region, exchange coupling and hardening occur between neighboring magnetically soft α-Fe and magnetically hard $Nd_2Fe_{14}B$ grains. It was proved that the presence of the intergranular layer benefits the acquisition of a magnet strongly coupled but remaining magnetically hardened [2], contrary to the expectation that such an intergranular phase may alleviate exchange interaction between hard magnetic and soft magnetic grains by separating them. An optimized intergranular region has a twofold function of enhancing exchange coupling between soft and hard phases [3,4] by developing a high degree of crystallographic coherence among adjacent grains, and quite oppositely, suppressing the averaging effect of anisotropy in hard grains to increase coercivity by detaching the grains [5]. Numerical simulation [6,7] with assumption of non-ideal grain boundaries indicates that a deviation of magnetic anisotropy constant K and exchange energy constant A at the intergranular regions from that of the basic structure has a direct influence on coercivity H_c and also reduced remanence m_r. It turns out that remanence and coercivity decrease approximately linearly with simultaneous reduction of exchange and anisotropy constants near grain boundaries. There are still much to be clarified and understood about functions of intergranular phase, such as what is the most appropriate intergranular structure and how it controls exchange coupling and hardening mechanisms amongst those neighboring soft and hard grains, and which heat treatment is the most appropriate to develop an optimal magnet.

These nanocomposite magnets are usually produced by the melting–spinning method directly with controlled cooling rates or by crystallizing melt–spun amorphous ribbons. In either quenching or annealing process, since initial and final phases do not have the same or similar concentration, grain growth will be continued only by long-range diffusion, which is hindered by the slow diffusion rates of certain species, like Nd, for instance [8]. When quenched, a frozen grain boundary structure in an as-spun nanocomposite is usually not maturely developed, due to the limited diffusion speed and time of the atoms, and it is prone to adjust itself to a more stable state afterward through stress releasing and to further short-range atom interdiffusing, even under room temperature conditions. The adjustment is most likely in the intergranular areas where different crystalline forefronts meet and insoluble impurities or unbalanced concentrations converge. Apparently, such adjustment will greatly alter features of the intergranular regions, resulting in significant changes in the interaction between neighboring grains, and in averaging effect of magnetic hardness to the hard grains. In this paper, effects of aging at room temperature on magnetic behaviors of $Nd_2Fe_{14}B$/α-Fe-based exchange-coupled powders are examined. Moreover, the functions of the intergranular region on the magnetic exchange hardening mechanism are also discussed.

2. Experiments

Ingots of Nd–Fe–B–X (X = Ti or In) composites were prepared by arc-melting in argon atmosphere (its purity is 99.9) with purities of starting elements: 99.8% of iron, 99.5% of neodymium, 99.9% of indium and 98% of $Fe_{1.3}B$ alloy and 99.9% of titanium, respectively. The ingots were broken and re-melted in an arc-melting crucible and poured onto a molybdenum wheel surface rotating at a speed from 5 to 25 m/s to form as-spun flakes. Thickness of the flakes was measured between 0.01 and 0.06 mm. Selected samples were heat treated in a quartz tube furnace in argon atmosphere at 200 °C for various time intervals from 0 to 9 h. Hysteresis loops were measured in a vibrating sample magnetometer (VSM) with a maximum operating field of 15 kOe. The X-ray diffraction (XRD) measurements with K_α of Cu were carried out on powder samples. Curie temperatures were measured in a thermal magnetometer with a fixed applied field of 1.6 kOe. Phase transition points were scanned up to 600 °C by using a differential thermal analyzer (DTA) with a scanning speed of 5 °C/min.