Journal of Magnetism and Magnetic Materials 411 (2016) 128–132

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Relationship of magnetic behavior and surface spin coupling in Hematite nanowires bundles

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ARTICLE INFO

Article history:
Received 28 September 2015
Received in revised form
18 January 2016
Accepted 15 March 2016
Available online 16 March 2016

Keywords:
Nanostructures
Magnetic materials
Chemical synthesis
Magnetic properties

ABSTRACT

Hematite (α-Fe_2O_3) nanowires were synthesized using mesoporous SBA-15 silica as the hard templates, and then the well-dispersed α-Fe_2O_3 nanowires (NW_S) were separated from the ordered α-Fe_2O_3 nanowires bundles (NWB_S) by the centrifugation technique. X-ray diffraction (XRD), transmission electron microscopy (TEM) and surperconducting quantum interference device (SQUID) were used to characterize the microstructure and magnetic properties of the as-prepared samples. All results indicated that the α-Fe_2O_3 NW_S and NWB_S with the different interwires distance presented the same diameter for nanowires, which was possible to reveal the exchange interaction between α-Fe_2O_3 NWBs. Both samples showed ferromagnetism and α-Fe_2O_3 NW_S presented superparamagnetism at high temperature. The magnetic results indicated the surface spin between the neighboring nanowires of α-Fe_2O_3 NWBs coupled each other. The saturation magnetization of α-Fe_2O_3 NWB_S was lower than that of α-Fe_2O_3 NW_S, while the coercivity and Curie temperature were larger. It was concluded that the surface spin coupling could weaken the surface effect on the magnetic properties for nanostructures.

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1. Introduction

Magnetic nanomaterials were of great interests for researchers in a wide range of fields, including magnetic fluids [1], catalysis [2,3], biotechnology/biomedicine [4–6], magnetic resonance imaging [7], data storage [8], and environmental remediation [9]. For nanoparticles, the magnetic properties not only depended on their intrinsic magnetism, but also were affected by the magnetic interaction between nanoparticles. The magnetic interaction was successfully applied on the spin valve and the double-phase permanent magnet of neodymium iron boron, which could greatly improve the performance of magnetic materials and devices [10–12]. More attention had been paid to the influence of the small size effect and the surface effects on the magnetic properties of nanoparticles, while the magnetic exchange interaction between nanoparticles were ignored. As the result, the magnetic interaction between nanoparticles was averagely thought as the magnetism of nanoparticles. With the different microstructure of nanoparticles, especially for the shape, size and interparticles distance, the researches of the magnetic interaction between nanoparticles were very difficult, which limited the development of the magnetic mechanism of nanoparticles [13,14]. Therefore, the synthesis of the monodisperse nanoparticles with the well-defined shape, a controlled composition and tunable interparticle separations was one of the most important tasks for the magnetic nanoparticles.

The uniform morphology and diameter was important for the study on the intrinsic magnetism of nanoparticles, which avoided the replacement the real magnetic parameters with the average value. Recently, the studies about the controllable nanoparticles had been quite mature, and many different methods had been developed for the synthesis of the magnetic nanoparticles, such as hydrothermal/solvothermal method [15,16], chemical deposition [17], coprecipitation method [18], template method [19] and so on. Bhattacharjee et al. [20] synthesized the monodispersed Co_3O_4 nanoparticles by a thermal decomposition method of size 4–12 nm. Yang et al. [21] synthesized the monodispersed Fe_2O_3 nanoparticles with the size around 10 nm via a precipitation method. Castellanos-Rubio et al. [22] synthesized the monodispersed Fe_3O_4 nanoparticles through the thermal decomposition method. Liu et al. [23] reported a new solvothermal method for the preparation of the magnetic well-controlled Fe_3O_4 nanoparticles with the size ranging from 7 nm to 12 nm.

The controllable microstructure (especially for interparticles distance) was very necessary to reveal the magnetic exchange interaction between the nanoparticles, which was very helpful to

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http://dx.doi.org/10.1016/j.jmmm.2016.03.055
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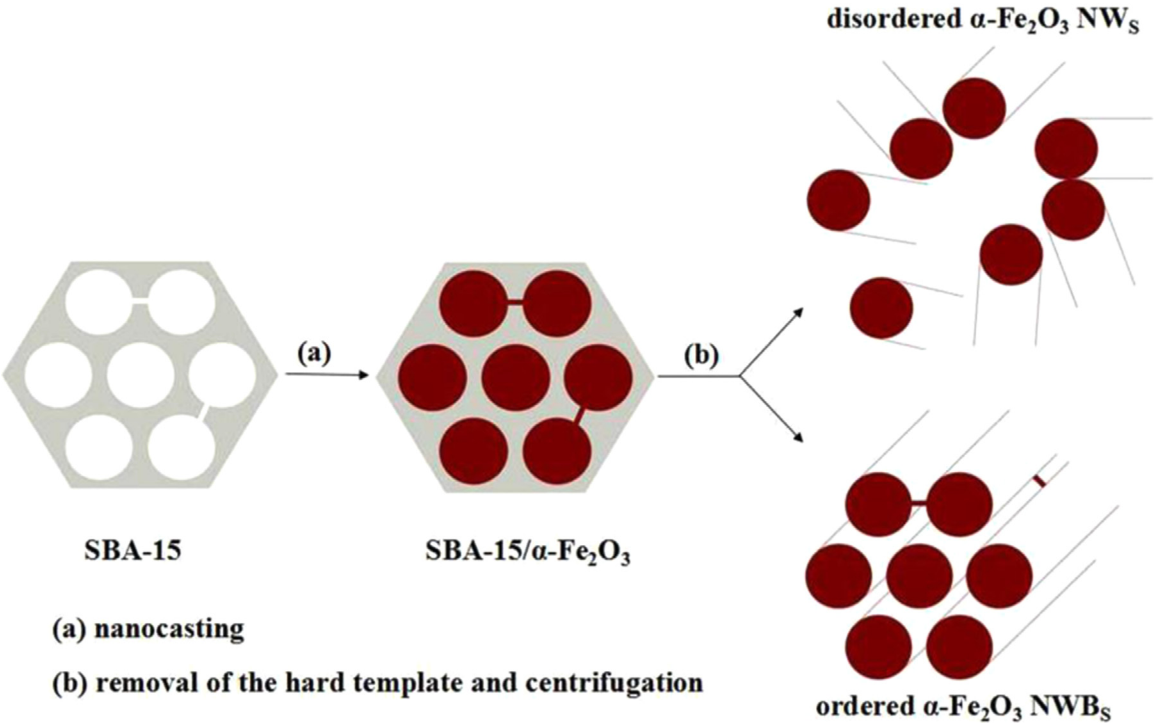

Fig. 1. The flow chart of the synthesis of α-Fe_2O_3 NW_S and NWB_S.

discuss the intrinsic magnetic properties of nanoparticles. The nanoparticles with a tunable interparticles distance should possess the 3D ordered packed microstructure, which was usually synthesized using the ordered mesoporous (micropores) materials as the hard templates. Since the microstructure of the hard templates could be adjusted with the chemical condition, the size and morphology of the guest magnetic nanoparticles could be tailored further. Ordered mesoporous materials with the tunable interpores separation, regular and narrow distribution pore and easily-eliminated skeletal structure, such as SBA-15, SBA-16, FDU-12, KIT-6 and the ordered mesoporous carbons, became the natural reactors to prepare the magnetic nanoparticles assemble [24–29]. This template-based nanocasting approach could also eliminate the aggregation of magnetic nanoparticles. The different kinds of magnetic nanoparticles with the same interparticles distance such as Cr_2O_3 [30,31], γ-Fe_2O_3 [32], Co_3O_4 [33,34] and so on, were synthesized by the nanocasting route with ordered mesoporous materials. Wang et al. [35] described that a series of magnetic γ-Fe_2O_3, Fe_3O_4, and Fe nanoparticles were successfully synthesized with the ordered mesoporous carbons as hard templates. In our previous works, the Co_3O_4 nanowires arrays with the same interwires distance were synthesized using SBA-15 silica as the template [36]. And 3D packed Co_3O_4 nanosphere assembles were prepared with LP-FDU-12 and the diameter and the interspheres distance could be tailed to some extent [37]. Furthermore, the uniform Fe_xO_y hollow nanospheres were also synthesized by the nanocasting pathway [38].

With the larger interparticles distance, the magnetic exchange interaction between nanoparticles could be easily distinguished from the intrinsic magnetic behavior of nanoparticles. Herein, the two kinds of α-Fe_2O_3 nanowires with the same diameter were synthesized with the much difference on the interwires distance, and then the magnetic interaction was discussed in detail. In this paper, α-Fe_2O_3 nanowires were synthesized with the nanocasting method, and then were divided into the well-dispersed α-Fe_2O_3 nanowires (NW_S) and ordered α-Fe_2O_3 nanowires bundles (NWB_S) by the centrifugation technique. After the extensive analysis of the magnetic behavior of the α-Fe_2O_3 NW_S, the magnetic properties of the α-Fe_2O_3 NWB_S were discussed in detail. Furthermore, the influence of the microstructure (especially for the interwires distance) on the magnetic properties was deduced. As the result, the relationship between the surface spin coupling and magnetic behavior of α-Fe_2O_3 NWB_S could be concluded.

2. Experimental section

All chemicals were of analytical grade and used as purchased without any further purification steps. Ordered mesoporous silica template SBA-15 was prepared by the method described previously [39]. 5.85 g of the surfactants P_{123} (poly (ethyleneglycol)-poly (propylene glycol)-poly (ethylene glycol)) was added to the mixture of 94.235 g of HCl (2 mol) and deionized water. The mixture was vigorously stirred for 2 h. Then, a total of 13.4 ml of tetraethyl orthosilicate (TEOS, 98%, Aldrich) as the silicon source was added and stirred for 5 min. After being aged at 45 °C for 24 h, the final mixture was transferred into a Teflon-lined autoclave, which was treated hydrothermally at 120 °C for 24 h before cooling to room temperature naturally. The product was filtered and washed with deionized water and ethyl alcohol, and then the white powder was calcined at 550 °C for 6 h to remove the $P_{123.}$

The preparation of α-Fe_2O_3 NWs and NWBs was performed as the following, which the flow chart was given in Fig. 1. Iron nitrate and SBA-15 powders (the atomic ratio of Si: Fe=2: 1) were dissolved in ethanol to make a solution and stirred to dry, then hexane was added until a fine powder formed. After the powder was heated at 550 °C for 6 h, 2 M NaOH aqueous solution was added to the above powder to remove the silica template. The α-Fe_2O_3 NW_S were separated from α-Fe_2O_3 NWB_S with the 13,000 rps by centrifugal separation technique. The two samples were filtered and washed with deionized water and ethyl alcohol, and then was dried at 80 °C for 4 h.

The phase compositions and microstructure of α-Fe_2O_3 nanowires were characterized by X-ray diffraction (XRD: XD-5A, Cu

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