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Reduction behavior of barium hexaferrite into metallic iron nanocrystallites

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Abstract

Barium hexaferrite ($BaFe_{12}O_{19}$) powder was prepared through the ceramic route by calcination of a stoichiometric mixture of barium carbonate and iron oxide at 1200 °C for 2 h. Compacts of synthesized $BaFe_{12}O_{19}$ powder were isothermally reduced in hydrogen atmosphere at 600–1000 °C. Based on thermogravimetric analysis, the reduction behavior of $BaFe_{12}O_{19}$ and reaction kinetics and mechanism were studied. The initial ferrite powder and the various reduction products were characterized by X-ray diffraction analysis,a scanning electron microscope and a vibrating sample magnetometer to reveal the effect of hydrogen reduction on composition, microstructure and magnetic properties. The reduction rate was found to be controlled by interfacial chemical reaction mechanism. Metallic iron was formed in nanosized regimes while grain growth and coalescence were observed at higher reduction temperatures. Magnetic measurements show diluted magnetic properties for the formed metallic iron, whereas coercivity values were in the range 14.1–55 Oe and saturation magnetization varied from 24.79 to 36.15 emu/g. These values have been ascribed to the formation and growth of nanosized metallic iron particles as the reduction treatment proceeded.

Keywords: Barium hexaferrite; Reduction; Nanocrystallite; Metallic iron; Diluted magnetic

1. Introduction

The synthesis of ultrafine magnetic powders for applications such as magnetic recording and magnetic fluids has received wide attention in recent years. Metallic iron particles have been used as magnetic media to prepare metal tapes due to their high magnetic moment and coercivity. Many investigators [1–3] studied the preparation of metallic ultrafine particles and its magnetic properties based on various techniques like laser-induced pyrolysis, mechanochemical processing, etc.

Carpenter [4] studied the preparation of stable magnetic nanoparticles composite based on iron. It was clearly found that these nanocomposite materials offer enhanced magnetic properties over other composites based on the presence of iron oxide. Uniform and stable α-Fe nanoparticles of around 40 nm in width and axial ratios from five to seven have been obtained from hematite (α-Fe_2O_3) without any additive by heating under a hydrogen atmosphere [5]. There is a minimum temperature (400 °C) and a minimum hydrogen flow (20 l/h) to reduce the hematite particles to metal in a reasonable time (4 h), preserving the size and the shape of particles. The main change in the material is related to the crystallite size of metal particles, which increases as reduction proceeds. The magnetic properties of nanoparticles compressed into pellets and dispersed in the composites were studied by Baker et al. [6] and Kuhn et al. [7]. The particles were observed to exhibit increased coercivity and exchange bias. The exchange bias was observed to increase with oxide shell thickness. The magnetism in nanoparticle composites was studied as a function of nanoparticle loading. It was observed that when particles were dispersed into the nanocomposite, coercivity was increased, suggesting a heightened anisotropy barrier. Similarly, the magnetic relaxation results indicate that the composites exhibit significantly reduced relaxations through the entire temperature range, as

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compared to the compressed pellet. This observation supports the possibility of heightened anisotropy barriers due to reduced dipolar interactions [6].

The reduction of ferrite compounds was rarely studied. The heat treatment in hydrogen or nitrogen atmosphere followed by the re-calcination process was handled. Ebrahimi et al. [8–12] studied the hydrogenation and re-calcination processes for synthesized strontium hexaferrite powder. It was found that Sr-hexaferrite decomposed into $Sr_7Fe_{10}O_{22}$ and Fe_2O_3,which reduced to Fe and FeO. A lower coercivity value is produced after hydrogen treatment while a higher value was achieved by re-calcination in air. The values of saturation and remanence magnetization after re-calcination are almost the same as those of the initial synthesized ferrite while only coercivity is increased. As reduction proceeded, a large number of fine sub-grains were formed with magnetic behavior approaching that of single domain material and so higher coercivity is obtained after re-calcination. Also, the optimum re-calcination temperature should be enough only for re-formation of the hexaferrite phase; otherwise, undesirable grain growth occurs at much higher temperature. In the presence of nitrogen gas atmosphere, a lower reduction degree was obtained that stopped at magnetite phase [9,10]. Due to the magnetically softer nature of the magnetite phase, a significant decrease in the intrinsic coercivity was detected. Also as in the hydrogen atmosphere, after re-calcination, higher values of M_s, M_r and H_c were obtained similar to that of the initial synthesized Sr-hexaferrite powder.

As mentioned above, the development of nanoparticle magnetic materials is very important and should be extensively studied. At the same time, reduction behavior of the ferrite compound and its kinetics reaction mechanisms are rarely handled and not covered at all in previous research work. From these points of view, the present investigation handled hydrogen reduction of barium hexaferrite ($BaFe_{12}O_{19}$) with a double target: to understand the reduction behavior and kinetics reaction mechanism of this ferrite type with its effect on composition, microstructure and magnetic properties, and to consider the reduction of $BaFe_{12}O_{19}$ as a source for the synthesis of ultrafine magnetic metallic iron powder.

2. Experimental

The ceramic route was applied for the preparation of $BaFe_{12}O_{19}$ powder. Stoichiometric ratios of analytical reagent-grade barium carbonate and ferric oxide were well mixed manually in agate mortar. The mixture was dried and then fired at 1200 °C for 2 h. An equal weight of the produced ferrite powder was compressed into briquettes of 1 cm diameters and 3 g weight that were dried at 105 °C.

The reduction of barium hexaferrite compacts was carried out at 600, 700, 800, 900 and 1000 °C in hydrogen gas atmospheres (100% H_2). The course of reduction was

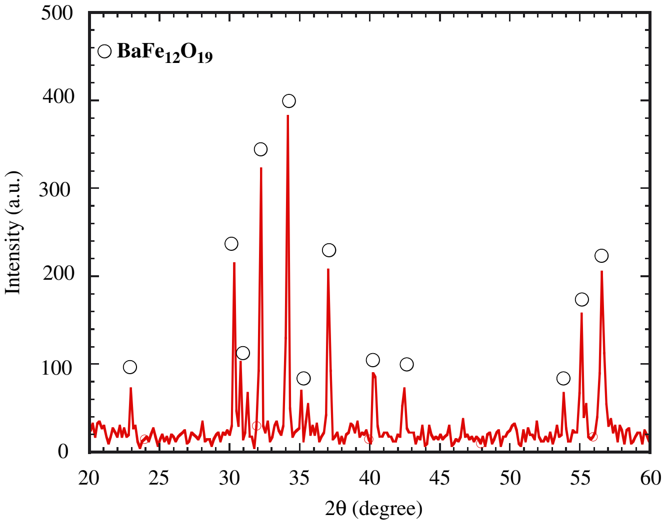

Fig. 1. XRD analysis of the synthesized barium hexaferrite (Ba $Fe_{12}O_{19}$) powder.

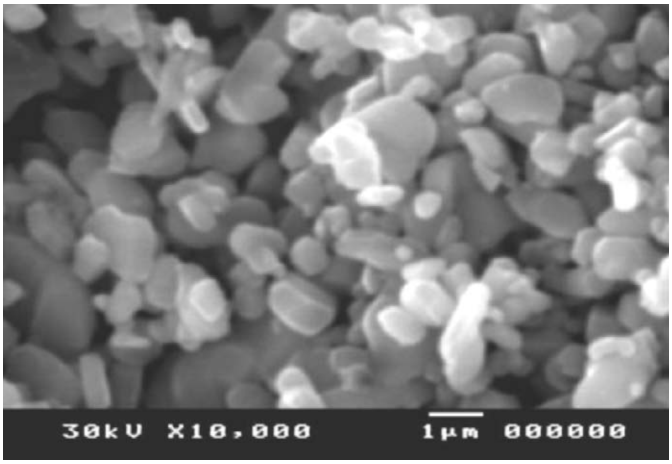

Fig. 2. SEM micrograph for the prepared ferrite powder.

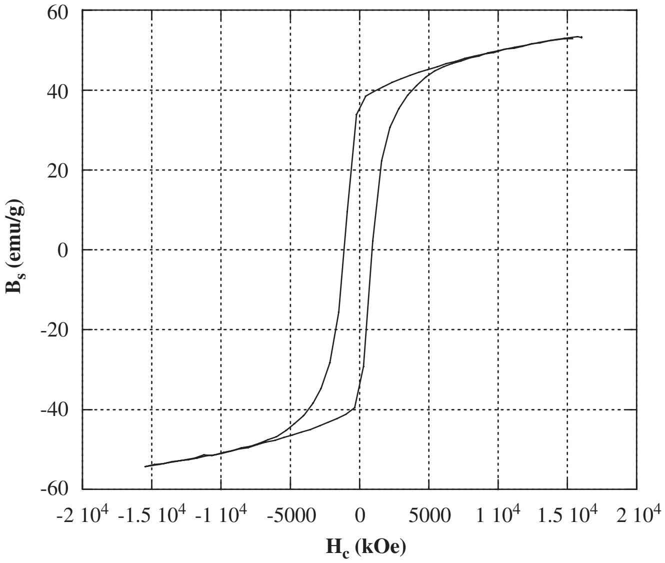

Fig. 3. *B–H* hysteresis loops for the synthesized ferrite powder.