



BaFe₁₂O₁₉ powder with high magnetization prepared by acetone-aided coprecipitation

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ABSTRACT

BaFe₁₂O₁₉ particles with high magnetization were produced using an acetone-aided coprecipitation process. An aqueous solution of iron and barium nitrates, in an Fe³⁺/Ba²⁺ molar ratio of 12, was added in a stirred precipitation liquid medium composed of H₂O, CH₃(CO)CH₃ and NH₄OH. After reacting metallic ions with ammonia, the precipitates were formed, centrifugally filtered, freeze dried and calcined. Effects of amount of the acetone in the precipitation liquid medium on the formation of crystalline BaFe₁₂O₁₉ were investigated. The presence of acetone in the precipitation liquid medium can greatly promote formation of the crystalline BaFe₁₂O₁₉ at temperature as low as 650 °C and can enhance magnetization of the derived particles. On the other hand, raising the calcination temperature can effectively accelerate development of crystallite morphology and magnetic characters of the barium hexaferrites. While the barium hexaferrite powder obtained without acetone additions and calcined at 1000 °C had magnetization (measured at 50 kOe; *M*(50 kOe)) of 63.5 emu/g, remanence magnetization (*M*_r) of 31.3 emu/g and coercivity (*H*_c) of 4.7 kOe, the single magnetic domain size BaFe₁₂O₁₉ powder with *M*(50 kOe) of 70.6 emu/g, *M*_r of 34.4 emu/g and *H*_c of 3.7 kOe was produced at 1000 °C, using a precipitation liquid medium of 64 vol% acetone.

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1. Introduction

Barium hexaferrite (BaFe₁₂O₁₉), as permanent magnet, is widely used in electronic industries due to its excellent magnetic properties, such as large magnetization (72 emu/g at 25 °C), high Curie temperature (450 °C), high magnetic anisotropy and high coercivity (6.7 kOe), as well as excellent chemical stability and corrosion resistivity [1–6]. The characteristics of BaFe₁₂O₁₉ particles are very important for manufacturing permanent ceramic magnets because of their influence on the quality of final products. To have a better performance, barium hexaferrite particles are required to be of single magnetic domain, good chemical homogeneity and narrow particle size distribution. Conventionally, solid-state reactions (or call ceramic methods), including milling and firing of the mixture of iron oxide and barium carbonate, are used to synthesize barium hexaferrite powder. The firing temperatures used are 1200 °C or above [7,8]. Although firing at high temperatures assures the formation of BaFe₁₂O₁₉ phase, larger particles (> 1 μm) are often obtained in this firing stage. Milling the solids usually leads to introduction of impurities into material compositions, generation of lattice strains in molecular structure and production of particles

with irregular shape. All of these will have negative impacts on developing microstructures of the ferrite products. Therefore, several nonconventional techniques, such as the glass crystallization method [9–12], the hydrothermal technique [13–17], the precursor method [18–22], the coprecipitation method [23–27] and others, have been used or are under development for preparing ultrafine barium hexaferrite particles. Among these, the coprecipitation method is one of the techniques suitable for the low-cost and mass production of barium hexaferrite particles.

In coprecipitation, the salts containing iron and barium elements are dissolved in a proper solvent to form a solution, followed by adding the precipitating agent to force the metallic cations in the solution to convert to solid phase(s), as precipitates. The solid precipitates were then calcined at predesigned temperature to form barium hexaferrite powder. Water, due to its ease of acquirement and non-toxicity, is usually used as a solvent to dissolve the metallic salts. The precipitating agent commonly used is ammonia (NH₄OH) or caustic soda (NaOH) since hydroxide ions easily attack cations to form metallic hydroxides, and most of metallic hydroxides have low solubility in water. However, the solubility of barium hydroxide in water has a finite value though iron (III) hydroxide is insoluble in water. To overcome this problem and ensure the precipitates containing the required amounts of Fe³⁺ and Ba²⁺, the aqueous solution of metallic salts containing Fe³⁺/Ba²⁺ molar ratios less than 12 (a stoichiometric ratio of

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$\text{Fe}^{3+}/\text{Ba}^{2+}$ to form $\text{BaFe}_{12}\text{O}_{19}$) is commonly adopted to prepare barium hexaferrites of high quality by coprecipitation [26–30].

In this study, a facile acetone-aided coprecipitation process was proposed to produce crystalline barium hexaferrite particles with high magnetic properties from an aqueous solution containing $\text{Fe}^{3+}/\text{Ba}^{2+}$ molar ratio of 12. Acetone was added in the precipitation liquid medium to adjust solubility of the derived metallic hydroxides during precipitation. Effects of the amount of acetone additions and the calcination temperature on characteristics and magnetic properties of the derived $\text{BaFe}_{12}\text{O}_{19}$ particles were investigated.

2. Experimental detail

2.1. Sample preparation

$\text{BaFe}_{12}\text{O}_{19}$ particles were synthesized using an acetone-aided coprecipitation method. Before reaction, two solutions (Soln-A and Soln-B) were prepared. Soln-A was formed by dissolving $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (99% purity, Showa) and $\text{Ba}(\text{NO}_3)_2$ (99% purity, Showa), in a stoichiometric ratio to form $\text{BaFe}_{12}\text{O}_{19}$, in distilled water. The total metallic concentration of Soln-A was controlled at 0.85 M. Soln-B, acting as precipitation liquid medium, was a solution composed of distilled H_2O , acetone ($\text{CH}_3(\text{CO})\text{CH}_3$; 99.7%, Echo) and ammonia solution ($\text{NH}_4\text{OH}_{(\text{aq})}$; 29 wt%, Fisher) with volumetric ratio of $\text{H}_2\text{O}:\text{CH}_3(\text{CO})\text{CH}_3:\text{NH}_4\text{OH}_{(\text{aq})}=x:y:250$. Soln-A of 110 mL was then slowly added to the stirred Soln-B of 690 mL to undergo coprecipitation reaction at 20 °C for 2 h. Table 1 shows the various compositions of reaction system and the corresponding specimen codes. The amount of acetone in Soln-B was varied from 0 to 64 vol%. After reaction, the pH of the liquid system was 10.8 ± 0.1 for all the cases. The resultant suspension was centrifugally filtered. The collected wet cake was frozen at -45 °C and then dried in vacuum using a shelf type freeze dryer (FD12-2S-6P, Kingmech, Taiwan). The dried precipitates so obtained were calcined, in air, at different temperatures for 5 h or 48 h.

2.2. Characterization

Thermal behavior of the dried precipitates was investigated using a thermal analyzer STA 499 F3 (Netzsch, Germany), which is capable of performing thermogravimetric analysis (TG) and differential scanning calorimetry (DSC) simultaneously for the same sample. The organic or inorganic functional groups that existed in the specimens were detected using an infrared spectrometer (IR; Magna-IR spectrometer 550, Nicolet, Madison, WI, USA). The crystalline phases and crystal structure existing in the specimen were examined by X-ray diffraction analysis (XRD; D8A, Bruker, Germany). The X-ray source was Cu-K α radiation ($\lambda=1.54056 \text{ \AA}$), powered at 40 kV and 40 mA, and XRD data were collected from

Table 1
Compositions of the reaction system to prepare barium ferrite powder.

Specimen code	Soln-A ^a (mL)	Soln-B			
		Distilled H ₂ O (x, mL)	Acetone (y, mL)	NH ₄ OH _(aq) ^b (mL)	Percentage of acetone (vol%)
A00	110	440	0	250	0
A16	110	330	110	250	16
A32	110	220	220	250	32
A48	110	110	330	250	48
A64	110	0	440	250	64

^a Aqueous solution of ferric nitrate and barium nitrate, in a stoichiometric ratio to form $\text{BaFe}_{12}\text{O}_{19}$, with total metallic ions concentration of 0.85 M.

^b 29 wt% $\text{NH}_4\text{OH}_{(\text{aq})}$.

25 to 45° (2θ) with a step size of 0.04° and a count time of 3 s. Based on the integrated peak intensities of the corresponding XRD patterns, mass ratios of different crystalline phases existing in the specimen were also estimated using a DIFFRAC^{plus} EVA software (Bruker, Germany). The molar ratio of Fe to Ba element existing in the specimens was measured using energy-dispersive X-ray spectroscopy (EDS; Bruker, Germany). Morphologies of the particles were observed using scanning electron microscopy (SEM; LEO 1530, Carl Zeiss, USA). The magnetic characteristics of some selected specimens were measured at room temperature using a superconducting quantum interference device magnetometer (SQUID; MPMS 7, Quantum Design, USA).

3. Results and discussion

3.1. Thermal behavior and phase development

By varying the amounts of acetone added in the coprecipitation procedure, five different dried precipitates (named A00, A16, A32, A48 and A64; refer to Table 1 for their exact contents) were obtained. Fig. 1 shows TG and DSC curves of these five different precipitates. For the dried precipitate obtained without any acetone addition (i.e., A00), two endothermic and two exothermic changes were detected during the course of heating. The first endothermic change at temperatures ranging from 45 °C to 150 °C was due to evaporation of the adsorbed water/acetone. A00 experienced the first exothermic change at 175–270 °C, which accompanied about 7.9% loss in weight. At 290–480 °C, the second endothermic change took place in the specimen, resulting in another 5.4% weight loss. Between 575 °C and 845 °C, the second exothermic change occurred and caused a slight decrease in weight (ca. 1.6 wt%). After adding acetone to the precipitation liquid medium, the weight loss of the specimen during heating

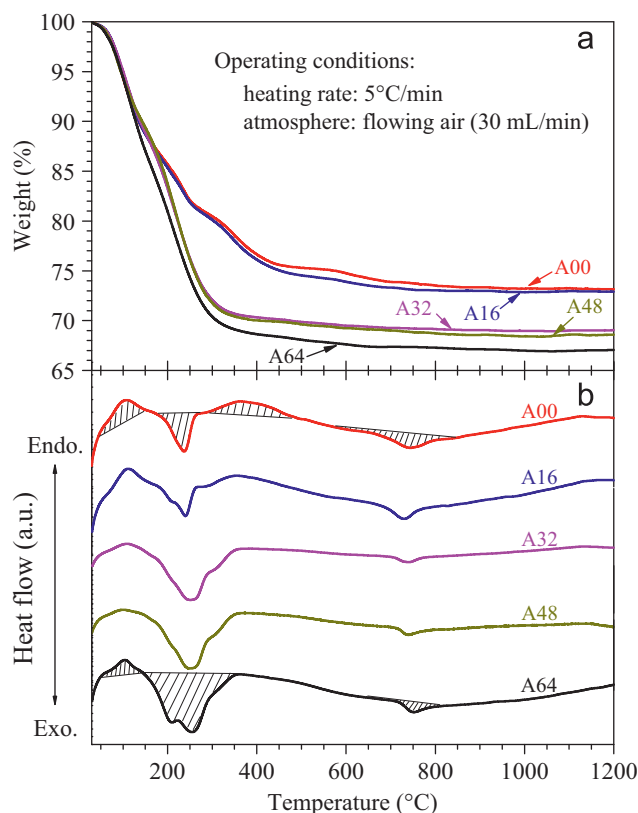


Fig. 1. (a) TG and (b) DSC curves of the dried precipitates.

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