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Optimization of Magnetic and Structural Properties of Sr Ferrite Prepared by Low-Temperature Combustion

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Fine strontium hexaferrite particles were prepared by the low temperature auto-combustion method. Aqueous solution of the corresponding metal salts and a combustible co-reactant compound — glycine were used as initial precursor. The resulting powders were calcined at various temperatures from 750°C up to 1100°C. The microstructure and magnetic properties were investigated by the Mössbauer spectroscopy, thermomagnetic analysis, scanning electron microscopy, and vibrating magnetometer. The particles of sizes less than 1 μm have monophasic hexagonal structure. Coercivity $H_c \approx 400$ kA/m, specific saturation polarization $J_{s-m} \approx 72 \times 10^{-6} \text{ T m}^3 \text{ kg}^{-1}$ and remanence polarization $J_{s-r} \approx 43 \times 10^{-6} \text{ T m}^3 \text{ kg}^{-1}$ were achieved for the Sr ferrite annealed at 850°C for 6 h.

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1. Introduction

The M-type strontium hexaferrite, which is a hard magnetic material, has been extensively used as a permanent magnet due to its low cost, relatively high coercivity, corrosion resistance, and chemical stability [1]. Magnetic properties of Sr or Ba ferrite powders strongly depend on the microstructure [2]. In order to get pure crystalline monodomain particles, various synthesis techniques have been developed, such as sol-gel technique [3], chemical coprecipitation [4], etc. A novel low-temperature combustion synthesis (LCS) is presented in this work, greater amount of the product can be obtained by this way.

Mixed solutions of metal nitrates and glycine were used to prepare ($\text{SrFe}_{12}\text{O}_{19}$). The effects of processing parameters, such as Sr/Fe ratio, nitrates/glycine ratio, reaction temperature and magnetic properties of the resultant Sr ferrite powders were investigated. Four samples prepared by various procedures were compared. The reproducibility of magnetic parameters was also an important criterion for procedure evaluation.

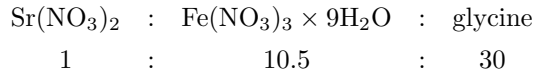
2. Experimental

$\text{Sr}(\text{NO}_3)_2$, $\text{Fe}(\text{NO}_3)_3 \times 9\text{H}_2\text{O}$ analytical reagent grade and $\text{HOOCCH}_2\text{-NH}_2$ (glycine) were used as raw components. Sufficient water was added to dissolve the

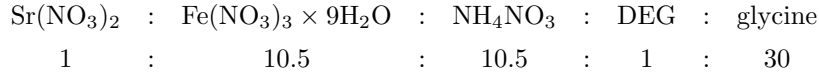
metal salts and glycine. Nitrate/glycine ratio considerably affects the combustion process and the grain size of prepared ferrite powder. The best product usually results when the glycine-to-cation molar ratio is adjusted to produce the largest bulk volume of ash [5]. The precursor mixture is processed by evaporating water into a glass-like substance. The auto-ignition was reached within a minute, yielding the brown-colored ash as a precursor. The ferrite particles generally grow in size during the calcination process in the laboratory muffle. Annealing temperatures 750, 800, 850, 900, 950, 1000, 1050, and 1100° were used for 6 hours. Ferrite particles calcined at 850° had typically submicron size with a narrow size distribution. In order to obtain a single phase Sr ferrite, optimal atomic ratio of Sr to Fe was chosen and appropriate amount of NH_4NO_3 and diethylglycole (DEG) into the precursor was added.

The microstructure and magnetic properties can be optimized by adding these modifiers and by correction of the calcination temperature.

The samples A and B were prepared in an amount of 10g of the final $\text{SrFe}_{12}\text{O}_{19}$ product. The molar ratio of sample A was chosen as follows:



Other ingredients except glycine was added into sample B in the following molar ratios:



The samples C and D were prepared in greater amount (approximately 100 g) in composition as sample B. The Mössbauer spectroscopy was carried out to study the cationic distribution on the hexagonal structure using a spectrometer with conventional constant acceleration mode and $^{57}\text{Co}/\text{Rh}$ γ -ray source. The spectra were fitted by NORMOS software. Temperature dependences of magnetic susceptibility $\chi(T)$ were measured by bridge method in an alternating magnetic field of 421 A/m at 920 Hz to obtain the Curie temperature T_c . The shape of these dependences gives also an information about the phase purity, presence of Hopkinson's peak gives an information about anisotropy and size of the particles. The magnetic properties of the ferrite powders were measured at room temperature using a vibrating sample magnetometer with an applied field of up to 755 kA/m. Scanning electron microscope (SEM) was used to examine the grain size and morphology.

3. Results

The Mössbauer spectra were analysed using five sextets. The Fe^{3+} ions in positions $4f_2$, $2a$ and $12k$ have octahedral locations, $4f_1$ tetrahedral and $2b$ occupy trigonal bipyramide. Hyperfine parameters of the Mössbauer spectra (B_{hf} and relative area R_A) at room temperature for the sample D annealed at 850 and 1050°C are compared with those of bulk sample (theoretical values [6]) in Table I. The relative area of each subspectrum is proportional to the number of ferric ions

on the corresponding sublattice. The magnetic properties of the samples *A*, *B*, *C*, *D* calcined at 850°C for 6 h were measured by a method described in [7], results are shown in Table II. All samples have promising values of magnetic parameters.

TABLE I

Influence of annealing temperature on hyperfine interaction parameters of the sample *D*.

Site	B_{hf} [T]			R_A [%]		
	850°C	1050°C	Theor.	850°C	1050°C	Theor.
$4f_2$	51.60	51.58	51.8	15.8	14.5	16.7
2a	50.86	50.85	50.6	7.4	9.2	8.3
$4f_1$	48.96	49.13	49.2	19.1	20.5	16.7
12k	41.33	41.35	41.3	50.0	48.9	50.0
2b	40.85	40.80	41.0	7.7	6.9	8.3

TABLE II

Magnetic properties of Sr ferrite powders.

Sample	J_{s-m} [10^{-6} T m ³ kg ⁻¹]	J_{s-r} [10^{-6} T m ³ kg ⁻¹]	H_c [kA m ⁻¹]
<i>A</i>	72.59	43.41	434
<i>B</i>	71.92	42.53	404
<i>C</i>	69.00	41.66	400
<i>D</i>	73.45	43.74	370

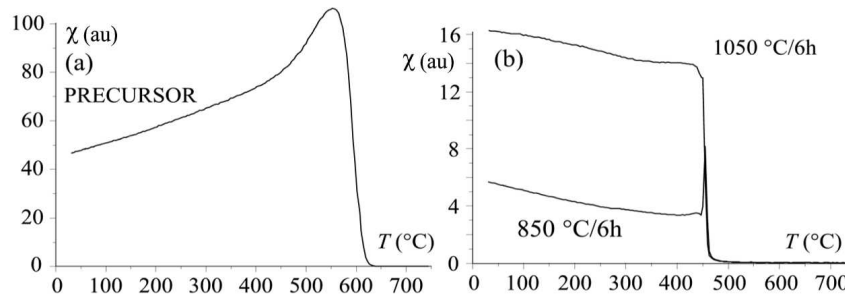


Fig. 1. Temperature dependence of magnetic susceptibility $\chi(T)$ of sample *D* – precursor (a), annealed at 850 and 1050°C (b).

Temperature dependence of magnetic susceptibility $\chi(T)$ of the sample *D* before calcination process (precursor) is shown in Fig. 1a. This dependence corresponds with the magnetized magnetite. The dependences for sample *D* annealed at 850 and 1050°C are shown in Fig. 1b. The $\chi(T)$ dependences exhibit a Hopkinson peak in the vicinity of the Curie temperature and confirms phase purity of ferrite powders. The height of the Hopkinson peak decreases with increasing annealing temperature, which is connected with grain size rising.

The morphology and grain size for sample *D* annealed at 850°C/6 h and 1050°C/6 h are shown in Fig. 2. The particle size of sample annealed at 850°C is much below 1 μm , while annealing at 1050°C yields particles of size over 1 μm of nearly hexagonal platelet shape.

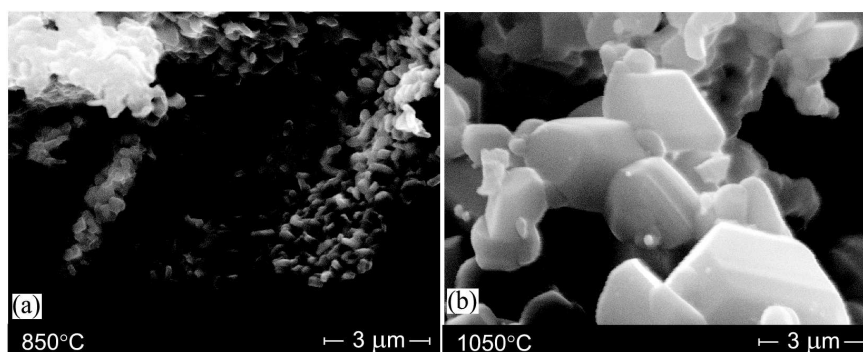


Fig. 2. Micrograph of particles size and morphology of sample *D*, annealed at 850°C for 6 h (a) and 1050°C for 6 h (b).

Fine strontium hexaferrite powders have been prepared at a relatively low calcination temperature by combustion synthesis also in greater amount of the product (over hundreds of grams). The initial nitrate/glycine ratio such as Fe/Sr ratio have a strong influence on the combustion process and the grain size of the product. The well-shaped $\text{SrFe}_{12}\text{O}_{19}$ particles were obtained at relatively low temperature 850°C for 6 h. It improved the magnetic properties coercivity around $H_c \approx 400$ kA/m for all samples (*A*, *B*, *C*, *D*). It is possible to use these materials as fillers in magnetocomposites.

Acknowledgments

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