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# Structural and magnetic properties of SrFe<sub>12</sub>O<sub>19</sub> hexaferrite synthesized by a modified chemical co-precipitation method

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## 1. Introduction

*M*-type strontium hexaferrite (SrFe<sub>12</sub>O<sub>19</sub>) was discovered in the 1950s by Philips' laboratories [1]. Due to appropriate magnetic properties, chemical stability, and low cost compared with rareearth compounds, it has attracted extensive interests in the past decades [2–4]. It is a hard magnet with high coercivity, which originates from high magnetocrystalline anisotropy with single easy magnetization axis. It has been recognized that it can be used as permanent magnets, recording media, telecommunication, and as components in microwave, higher-frequency, and magneto-optical devices [5–11].

The structure of *M*-type hexagonal is stacked alternatively by spinel ( $S = Fe_6O_8^{2+}$ ) and hexagonal ( $R = MFe_6O_{11}^{2-}$ ) layers. The  $O^{2-}$  ions exist as close-packed layers, with the  $M^{2+}$  substituting for an  $O^{2-}$  in the hexagonal layer. The  $Fe^{3+}$  ions are distributed in the five interstitial crystallographic sites of the close-packed layers, i.e. three octahedral (2a, 12k and 4f<sub>2</sub>), one tetrahedral (4f<sub>1</sub>), and one trigonal bipyramid (2b). The three parallel (2a, 12k and 2b) and two antiparallel (4f<sub>1</sub> and 4f<sub>2</sub>) sub-lattices, which are coupled by

#### ABSTRACT

*M*-type strontium hexaferrite (SrFe<sub>12</sub>O<sub>19</sub>) particles had been prepared by a modified chemical coprecipitation route. Structural and magnetic properties were systematically investigated. Rietveld refinement of X-ray powder diffraction results showed that the sample was single-phase with the space group of P6<sub>3</sub>/mmc and cell parameter values of a = 5.8751 Å and c = 23.0395 Å. The results of fieldemission scanning electronic microscopy showed that the grains were regular hexagonal platelets with sizes from 2 to 4 µm. The composition determined by energy dispersive spectroscopy is the stoichiometry of SrFe<sub>12</sub>O<sub>19</sub>. The ferrimagnetic to paramagnetic transition was sharp with Curie temperature  $T_C = 737$  K, which further confirmed that the samples were single phase. However, it was found that the coercivity, saturation magnetization and the squareness ratio of the synthesized SrFe<sub>12</sub>O<sub>19</sub> samples were lower than the theoretical values, which could be explained by the multidomain structure and the increase of the demagnetizing factor.

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superexchange interactions through the  $O^{2-}$  ions, form the ferrimagnetic structure [12–14]. A schematic *M*-type structural representation and the surroundings of five Fe<sup>3+</sup> sites are shown in Fig. 1 by Collomb et al. [15].

Because the magnetic properties of  $SrFe_{12}O_{19}$  strongly depend on the size and shape of the particles [16,17], several routes have been used to prepare strontium ferrite, including the traditional sol-gel process [18,19], the solid-state method [17,20], the saltmelting method [21], ball milling [22,23], self-propagating hightemperature synthesis [24], and the chemical co-precipitation method [25].

As it is well known, the chemical co-precipitation method is usually used to synthesize magnetic oxides due to its simplicity and good-control of grain size. However, in previous reports it was always observed that there exist some undesirable intermediate non-ferromagnetic phases, which lead to poor magnetic properties and irregular shape for the derived  $SrFe_{12}O_{19}$  particles [26]. In this paper, single-phase  $SrFe_{12}O_{19}$  particles with relatively homogeneous size are successfully prepared by means of a modified chemical co-precipitation method. In the processing, ammonium oxalate monohydrate was used as a precipitator instead of alkali in order to control effectively the nucleation and growth of  $SrFe_{12}O_{19}$  particles. As a result, the grain size, shape and size distribution are well controlled.

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## 2. Experimental

Micron-sized SrFe<sub>12</sub>O<sub>19</sub> particles were prepared by the chemical co-precipitation method. All starting precursors were of highpurity compounds. First, Sr(NO<sub>3</sub>)<sub>2</sub> and Fe(NO<sub>3</sub>)<sub>3</sub> · 9H<sub>2</sub>O with Sr:Fe atomic ratio of 1:12 were dissolved in deionized water by gentle heating. Then, the aqueous mixture was slowly poured into (NH<sub>4</sub>)<sub>2</sub>C<sub>2</sub>O<sub>4</sub> · H<sub>2</sub>O solution and stirred for several minutes using a magnetic stirrer. The gelatinous precipitate was filtered and washed several times using deionized water until the pH value of the solution became neutral. The dried powders were sintered at 500 °C for 8 h in air, and then sintered at 1100 °C for 8 h in air. Finally, the obtained powders were pressed into pellets with  $\Phi$ 10 × 2 mm<sup>2</sup> under a pressure of 20 MPa and sintered at 1000 °C for 10 h in air. The crystal structure was examined by Philips X'pert PRO X-ray diffractometer (XRD) with  $CuK_{\alpha}$  radiation at room temperature. The morphology of grains was investigated by field-emission scanning electronic microscopy (FE-SEM), and the compositions were examined by the energy dispersive spectroscopy (EDS) in FE-SEM. Magnetization measurements from room temperature to 800 K were performed using a vibrating-sample magnetometer (VSM) accompanied by a quantum design physical properties measurement system (PPMS) (1.8 K  $\leq$  T  $\leq$  1000 K, 0 T  $\leq$  H  $\leq$  9 T).

# 3. Results and discussion

The room-temperature powder XRD pattern and Rietveld refinement of  $SrFe_{12}O_{19}$  sample are shown in Fig. 1. The good



Fig. 1. The crystal structure sketch map of the hexagonal M-type phase and the five Fe sites with their surroundings are displayed, which is taken from Ref. [15].



**Fig. 2.** The output from the Rietveld refinement analysis of the XRD pattern for  $SrFe_{12}O_{19}$  sample. The plus signs show raw experimental data, and the continuous line overlapping them refers to the calculated data. The vertical bars are the expected Bragg reflection positions. The difference between the experimental data and the calculated data is shown at the bottom.

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