

RESEARCH ARTICLE

Variation of magnetic properties in strontium magnet powders with Sr/Fe ratio

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Abstract

The aim of our current study was to investigate the impact of Sr/Fe ratio on both the phase formation and magnetic properties of strontium hexaferrite powders. To achieve this, we employed mechanochemical synthesis to synthesize the powders, followed by a heat treatment process at 950°C for a period of 1 hour under an open atmosphere. XRD patterns revealed that the primary phase in all powders was identified as the SrFe₁₂O₁₉ phase. The SEM image showed that the particles had a round morphology with partial agglomeration due to the reduction in particle size. The magnetic properties were found to be affected by Sr/Fe ratio. The (BH)_{max} values increased from 0.35 MGOe to 3.11 MGOe as the Sr/Fe increased from 1:11.0 to 1:12.0. The presence of secondary phases such as α-Fe₂O₃ and SrO weakened the magnetic properties. The weight fraction of the SrO phase increased with a decrease in stoichiometry from 1:12 to 1:11, which correlated with the weakening of the magnetic properties. The study concludes that the Sr/Fe ratio between Sr/Fe is a critical parameter in determining the magnetic properties of strontium hexaferrite powders.

1. Introduction

Strontium hexaferrite (SrM), also known as SrFe₁₂O₁₉, is a type of magnetic material that belongs to the family of ferrites. It is a highly anisotropic material with high magnetic anisotropy and exhibits excellent magnetic properties such as high coercivity, high magnetic moment, and low magnetic damping [1-3]. These properties make it an attractive material for various magnetic applications, including data storage, magnetic sensors, microwave devices, and permanent magnets [4, 5].

Strontium hexaferrite magnets are composed of a combination of strontium sulfate and iron oxide, which are sintered at high temperatures to form a ceramic material. The unique crystal structure of strontium hexaferrite gives it its magnetic properties. It has a hexagonal crystal structure, with a space group of P6₃/mmc, and consists of layers of iron oxide with strontium ions in between [6]. The layers are stacked in a way that creates a uniaxial magnetic anisotropy, which makes strontium hexaferrite magnets highly resistant to demagnetization [7].

Strontium hexaferrite magnets are commonly used in various applications that require strong permanent magnets, such as speakers, electric motors, generators, and magnetic separators. They are also used in microwave devices such as circulators, isolators, and phase shifters, where their low magnetic damping and high coercivity are advantageous [8, 9]. Furthermore, they are used in magnetic recording media such as magnetic tapes and hard disk drives due to their high magnetic moment and thermal stability [10].

The basic production process for sintered strontium hexaferrite magnets involves the traditional powder metallurgy method. In this method, different types of strontium hexaferrite powders produced using various techniques are subjected to

classification, pressing, and sintering processes to produce the sintered magnets. The techniques used to produce SrM magnet powders include traditional ceramic methods, mechanochemical synthesis, self-propagating high-temperature synthesis, co-precipitation, sol-gel, hydrothermal synthesis, spray pyrolysis, molten salt and Pechini methods [11-16]. These methods involve different chemical and physical processes to produce strontium hexaferrite powders, which are then used to manufacture high-quality sintered magnets for various industrial applications [17]. Tiwary et al. [18] utilized celestite and iron ore powders in their research for the preparation of strontium hexaferrite powder. The powder produced through the annealing process was sintered at 1250 °C for 1 hour to produce SrM magnets. As a result of these experimental studies, the maximum coercivity was determined to be approximately 270 kA/m (3390 Oe) after a synthesis time of 50 hours. In their research, Tiwary and Narayan utilized celestite (natural strontium ore) and tuff as raw materials for the preparation of strontium hexaferrite powder. They stated that a single-phase structure was achieved through 40 hours of mechanochemical synthesis, followed by annealing at 900 °C. The magnetic properties obtained for this synthesis time and annealing temperature were as follows: 2533 Gauss remanence, 3160 Oe coercivity, and 1.19 MGOe energy product [19]. F. Sánchez-De Jesús and colleagues produced M-type strontium hexaferrite from mechanically activated strontium and iron oxides through a high-energy milling process for 5 hours. The aim of their study was to investigate the differences between the traditional heat treatment and spark plasma sintering (SPS) methods in sintering the produced hexaferrite powders at relatively low temperatures (900 °C). The magnetic properties of the hexaferrites produced using both methods varied between H_c = 288-509 kA/m and B

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= 0.29-0.32 T. They stated that these magnetic properties were consistent with the permanent magnet values of nanostructured $\text{SrFe}_{12}\text{O}_{19}$ synthesized using other methods [20]. Luo prepared Sr-ferrite powders using SrCO_3 and Fe_2O_3 as raw materials through a mechanochemical synthesis process. The saturation magnetization of the annealed powders was measured to be 58.2 Am²/kg at room temperature, with a coercivity value of 281.2 kA/m. Luo stated that compared to the traditional ceramic method, the mechanochemical method provided the advantage of achieving a higher coercive field (coercivity) [21].

The objective of this study was to utilize the mechanochemical synthesis (MCS) production technique for the production of strontium hexaferrite magnets with a hexagonal crystal structure. By employing the MCS method, SrM powders were generated by altering the Sr/Fe ratio using SrSO_4 and Fe_2O_3 . The Sr/Fe ratio was systematically varied from 1:11 to 1:12.0 with a 0.1 increment in order to investigate the influence of this ratio on the magnetic properties. The findings of the study demonstrated that the magnetic properties of the SrM magnets were notably influenced by the Sr/Fe ratio.

2. Materials and methods

Strontium hexaferrite ($\text{SrFe}_{12}\text{O}_{19}$) particles were produced by utilizing SrCO_3 as the source of Sr and mill scale, a by-product of mild steel production, as the source of Fe. Initially, the mill scale was purified by washing it with distilled water and then drying it at 100°C for 8 hours. Next, the milled mill scale was heat-treated at 900°C for 2 hours in atmospheric conditions to convert FeO and Fe_3O_4 phases to Fe_2O_3 . The Sr/Fe amounts of Fe_2O_3 and SrCO_3 were mixed and subjected to a mechanochemical synthesis process through high-energy ball milling for 20 minutes. The SrCO_3 and Fe_2O_3 powder mixture had a different Sr/Fe ratio range of 1:11 to 1:12.

The SrCO_3 and Fe_2O_3 powders, along with 10 mm WC balls and 1.2 ml of ethanol as a process control agent, were combined and loaded into a 250 ml WC milling jar. The milling process was performed in air at ambient temperature, using a fixed rotating speed of 420 rpm and a ball to powder weight ratio of 10:1 for a duration of 25 minutes. Following the high-energy ball milling process, the synthesized powders were heat-treated at 950 °C for 1 hour in order to stabilize their crystalline structure, which may have degraded during the mechanochemical synthesis process. Zeiss brand LS 10 model scanning electron microscope (SEM) was used to morphological analysis. The crystalline phases of the samples were identified through XRD measurements using a PANalytical X'pert Powder³ X-ray diffractometer. The patterns were collected in the range of 20-60° and a time per step of 180. The percentages of different phases of the powders were determined using Rietveld refinement analysis. Magnetization studies were conducted at room temperature using a LDJ Electronics 9600 vibrating sample magnetometer (VSM).

3. Results and discussion

SEM image (in Figure 1) revealed that the particles synthesized using a 1:12.0 Sr/Fe ratio and a 200-minute mechanochemical synthesis process displayed a round morphology, with some partial agglomeration observed due to the reduction in particle size. Most of the particles were around 1 μm in size, with only a small number of particles larger than 3 μm detected. The XRD patterns of $\text{SrFe}_{12}\text{O}_{19}$ powders synthesized with Sr/Fe ratios ranging from 1:11 to 1:12.0, with an increment of 0.2, are shown in Figure 2. The primary phase

in all powders was identified as the $\text{SrFe}_{12}\text{O}_{19}$ phase by their XRD patterns, and the phase peaks of $\text{SrFe}_{12}\text{O}_{19}$ belonged to the hexagonal magnetoplumbite structure with a space group of P63/mmc, as determined by HighScore Plus software. The lattice parameters of the hexagonal crystal structure were calculated to be $a=b=5.88 \text{ \AA}$ and $c=23.06 \text{ \AA}$ for the 1:12 Sr/Fe ratio. The increase in the c lattice parameter is attributed to the presence of elements in the mill scale structure.

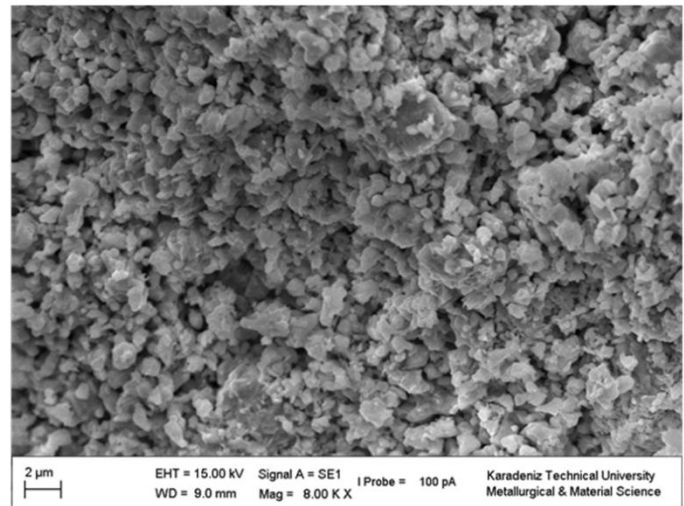


Figure 1. SEM image of strontium hexaferrite ($\text{SrFe}_{12}\text{O}_{19}$) powders after 200 minutes of mechanochemical synthesis

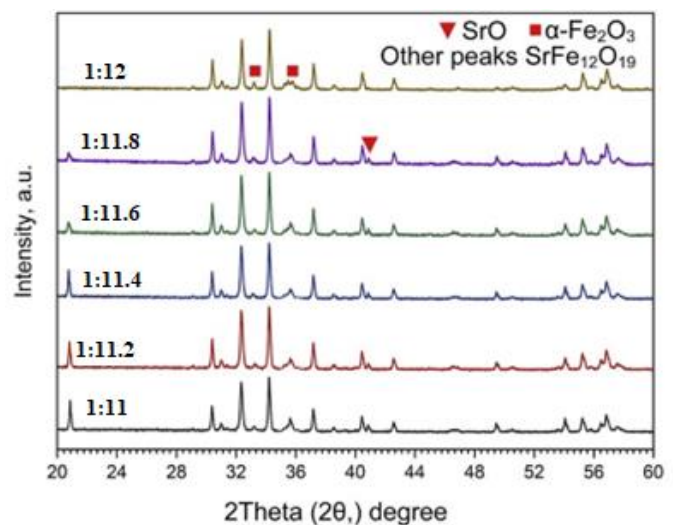


Figure 2. X-ray diffraction patterns (XRD) of mixtures with different Sr:Fe ratios

Strontium hexaferrite ($\text{SrFe}_{12}\text{O}_{19}$) is a well-known hard magnetic material with industrial applications, such as in magnetic data storage devices, microwave devices, and sensors. In this study, the effects of Sr/Fe ratios on the phase formation and magnetic properties of strontium hexaferrite powders synthesized by synthesis and heat treated at 950°C for 1 hour under an open atmosphere were investigated.

The results showed that the Sr/Fe ratio between SrCO_3 and Fe_2O_3 had a significant effect on the phase formation and magnetic properties of the synthesized strontium hexaferrite powders. When the Sr/Fe ratio was below the theoretical value of 1:12, $\alpha\text{-Fe}_2\text{O}_3$ and SrO phases formed in addition to the $\text{SrFe}_{12}\text{O}_{19}$ phase. The weight fraction of the SrO phase was

found to increase with a decrease in stoichiometry from 1:11.8 to 1:11, which explained the weakening magnetic properties.

The hysteresis loops of the synthesized powders were obtained, and magnetic properties such as coercivity (H_c), magnetic flux density (B_s), residual magnetic flux density (B_r), and maximum energy product ($(BH)_{max}$) were calculated. The results showed that as the Sr/Fe ratio increased from 1:11 to 1:12.0, the values of H_c , B_s , and B_r also increased. The $(BH)_{max}$ values, which are an indicator of the strength of a magnet, were obtained as 3.11, 2.36, 2.35, 0.4, 0.4, and 0.35 MGOe, respectively. It is noteworthy that the formation of the SrO phase increases as the Sr/Fe ratio decreases from 1:12 to 1:11. The weight fraction of this phase was found to increase from 1.6% to 7.4%, which correlates with the observed weakening of the magnetic properties. It is likely that the presence of the SrO phase interferes with the formation and alignment of the hard magnetic phase, resulting in weaker magnetic properties.

The formation of a "kink" in the hysteresis loop was observed in almost all samples, which was particularly prominent in the sample with a Sr/Fe ratio of 1:11. This shape distortion was reported to be due to the presence of secondary phases such as α - Fe_2O_3 and SrO, which are not the hard magnetic phase ($SrFe_{12}O_{19}$) present in the structure, or differences in particle size. Kink formation due to transitions between hard and soft magnetic phases or paramagnetic phases in the structure of any magnetic material was also observed. In conclusion, the Sr/Fe ratio between $SrCO_3$ and Fe_2O_3 plays a critical role in the phase formation and magnetic properties of strontium hexaferrite powders synthesized by mechanochemical synthesis and heat-treated at $950^\circ C$ for 1 hour under an open atmosphere. The results showed that as the Sr/Fe ratio increased, the magnetic properties improved, while the presence of secondary phases such as α - Fe_2O_3 and SrO weakened the magnetic properties. These findings can be useful for the optimization of the synthesis conditions and the development of high-performance magnetic materials.

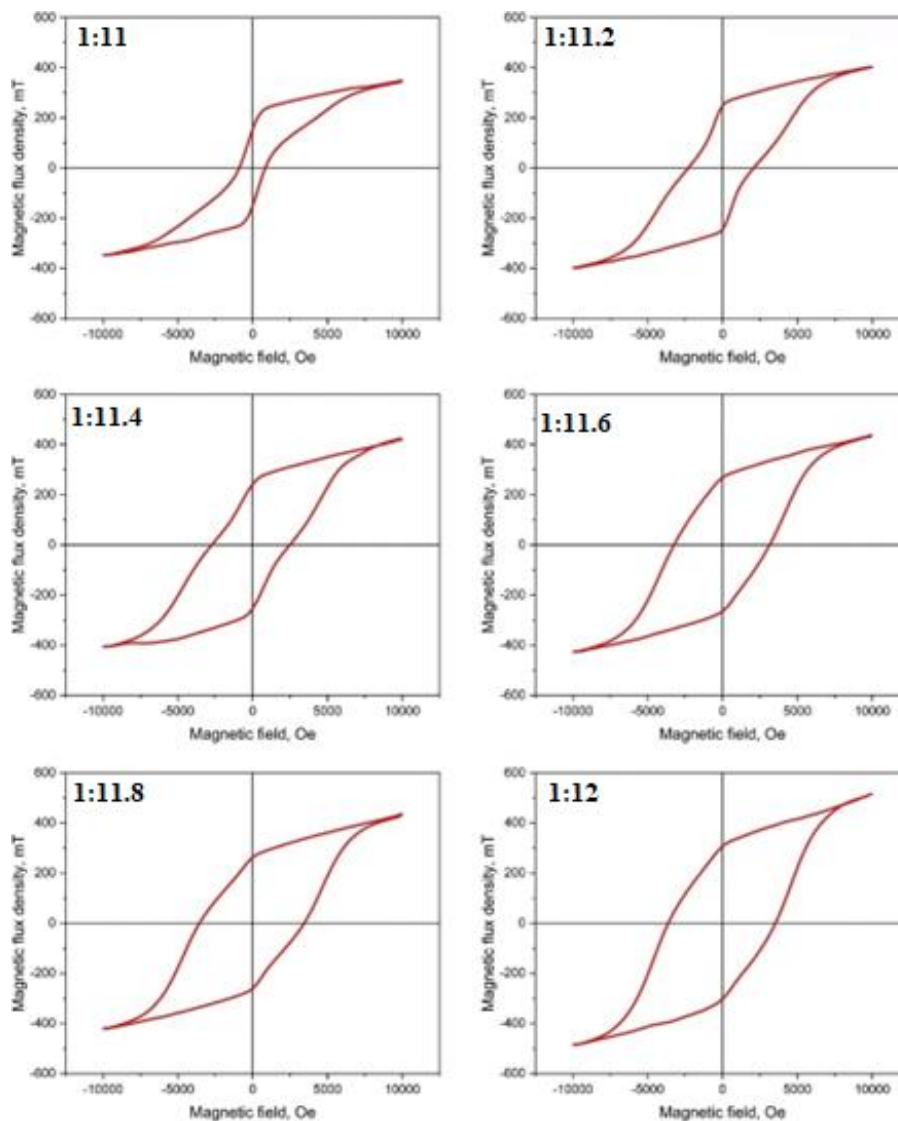


Figure 3. Magnetic hysteresis loops of produced hexaferrite powders

4. Conclusions

Based on the results of this study, it can be concluded that the Sr/Fe ratio between $SrCO_3$ and Fe_2O_3 is a critical parameter in determining the phase formation and magnetic properties of

strontium hexaferrite powders synthesized by mechanochemical synthesis and heat-treated at $950^\circ C$ for 1 hour under an open atmosphere. The XRD patterns revealed that the primary phase in all powders was identified as the $SrFe_{12}O_{19}$ phase with a

hexagonal magnetoplumbite structure. The SEM image showed that the particles had a round morphology with partial agglomeration due to the reduction in particle size.

The magnetic properties, such as coercivity (H_c), magnetic flux density (B_s), residual magnetic flux density (B_r), and maximum energy product ($(BH)_{max}$) of the synthesized powders were found to be affected by the Sr/Fe ratio. The $(BH)_{max}$ values increased from 0.35 MGOe for the sample with a Sr/Fe ratio of 1:11 to 3.11 MGOe for the sample with a Sr/Fe ratio of 1:12.0. The presence of secondary phases such as α - Fe_2O_3 and SrO weakened the magnetic properties, as evidenced by the kink formation in the hysteresis loops. The weight fraction of the SrO phase was found to increase with a decrease in stoichiometry from 1:12 to 1:11, which correlated with the weakening of the magnetic properties.

In summary, the optimization of the Sr/Fe ratio can lead to the development of high-performance magnetic materials with improved magnetic properties. These findings can be useful for the design and synthesis of strontium hexaferrite powders for various industrial applications, such as in magnetic data storage devices, microwave devices, and sensors.

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Author contributions

Sümran Bilgin: Conceptualization, Data curation, Investigation, Methodology
Sultan Öztürk: Project administration; Resources; Supervision

Kürşat İçin: Validation; Visualization; Roles/Writing-original draft; Writing - review & editing

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