



Effect of lanthanum substitution on dielectric relaxation, impedance response, conducting and magnetic properties of strontium hexaferrite



Basharat Want*, Bilal Hamid Bhat, Bhat Zahoor Ahmad

Solid State Research Lab, Department of Physics, University of Kashmir, Srinagar 190006, India

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ABSTRACT

Lanthanum strontium hexaferrite $\text{Sr}_{1-x}\text{La}_x\text{Fe}_{12}\text{O}_{19}$ ($x = 0, 0.08, 0.13, 0.18$) has been successfully synthesized by using citrate-precursor method and characterized by different techniques. The X-ray diffraction results revealed that the sample is crystalline in nature and is of single phase with the space group $P63/mmc$. The dielectric, conducting and impedance related studies have been carried out as a function of frequency and concentration of lanthanum in the frequency ranges of 20 Hz–3 MHz. Impedance studies were performed in the frequency domain to distinguish between bulk and grain boundary contributions of the material to the overall dielectric response. The electric response of the material was also modeled by an equivalent circuit and different circuit parameters were calculated. Magnetic characterization of the material was also performed and the effect of lanthanum concentration was studied. The hysteresis loop obtained from the magnetometer showed that with the increase of lanthanum concentration, the saturation magnetisation decreases while as coercivity increases.

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1. Introduction

Ferrite materials have always been a very important area of research due to their wide range of technological applications. Interest in the development of new ferrites has been rapid as compared to other areas of research [1–3]. Strontium hexaferrites ($\text{SrFe}_{12}\text{O}_{19}$) have attracted even more attention and became a subject of extensive study and interest [4]. Because they have good thermal durability, perfect chemical stability, unique magnetic and electrical properties and corrosion resistivity, they are used extensively in recording devices, telecommunication, magneto-optical and microwave devices [5,6]. These applications range from microwave to radio frequencies. So study of their dielectric behaviour at different frequencies is very important [7,8]. Ferrites have a very low conductivity. The order of magnitude of the conductivity greatly influences the dielectric and magnetic behaviour of ferrites. This aroused considerable interest in the electrical conductivity and the frequency dependent dielectric behaviour of ferrites [9–11]. The dielectric properties of ferrites depend upon several factors such as method of preparation, chemical composition and grain structure or size. Further, composition and frequency dependent dielectric studies of ferrites give valuable information about

conduction phenomenon in the ferrites based on localized electric charge carriers [12].

Keeping technological importance of hexaferrites into consideration, a number of cation substitutions in strontium hexaferrite has been done in order to enhance the saturation magnetisation or coercivity [13,14]. It has been observed that the coercivity improves upon La–Co substitution in strontium hexaferrite as compared to pure strontium hexaferrite [14]. The physical properties of ferrites have been found to be influenced by several factors such as the distribution of cations among the sublattices, nature of grain, grain boundaries, voids, and inhomogeneities [15]. So, the information about these parameters is very important in order to understand the behaviour of the material.

Impedance spectroscopy is a very important tool used for investigating the dielectric relaxation processes in polycrystalline samples. It is established that the complex impedance spectroscopy is a powerful tool to analyze the relationship between microstructure and properties [16]. The aim of the impedance measurements is identification of the physical processes and the determination of various electrical properties appropriate for the electrical system under study. The basic information about the dielectric properties of materials can be obtained from the complex impedance analysis. It can be used to separate several contributions of total impedance, arising from the bulk conductance and interfacial phenomenon viz. grain, grain boundary and other electrode interface effects [17]. The commonly used models for the impedance spectroscopy are

* Corresponding author. Tel.: +91 194 2420078; fax: +91 194 2421357.

E-mail address: bawant@kashmiruniversity.ac.in (B. Want).

electrical equivalent circuits consisting of resistors, capacitors, inductors and specialized distributed elements and give an insight into the physical process occurring inside the sample [18]. Ferrites are mostly studied because of their magnetic behaviour. Less attention has been paid to the study of their dielectric, impedance and conducting behaviour [19]. The objective of the present work is to study the effect of lanthanum doping on the dielectric properties, conductivity and impedance of strontium hexaferrite over a wide range of frequencies at room temperature. The magnetic behaviour of strontium hexaferrite was also studied at different lanthanum concentrations.

2. Experimental procedure

The polycrystalline samples of lanthanum substituted strontium hexaferrite $\text{Sr}_{1-x}\text{La}_x\text{Fe}_{12}\text{O}_{19}$ (where $x = 0.08, 0.13, 0.18$) were prepared through citrate-precursor method [20] using analytical reagents namely strontium nitrate anhydrous, ferric nitrate nonahydrate, lanthanum nitrate hexahydrate, citric acid anhydrous and ammonia (25%). The ratio of citric acid and metal nitrates was fixed at 1:1 and pH was maintained at 6.5. The powder was ground in motor pestle for 1 h and then kept at 950 °C for 3 h. The sintered samples were then used for various characterizations. X-ray powder diffraction data of the present samples were collected at room temperature with Bruker AXS D8 Advance powder diffractometer using $\text{Cu K}\alpha$ ($\lambda = 1.5406 \text{ \AA}$) radiation and an applied voltage and current of 40 kV and 35 mA, respectively. The data were recorded in the range of $20^\circ \leq \theta \leq 80^\circ$ with a step scan rate of 0.020° per step and a count time of 31.2 s per step. The micro structural studies were made by using the JEOL JSM-6390LV scanning electron microscope. The method of energy dispersive analysis of X-rays was carried out on the samples by using a JEOL JED-2300 energy dispersive spectrometer.

The dielectric measurements were carried out on pellets, obtained by finely grinding the samples and pressing the resulting powder under a pressure of 150 kg/cm² using a hand operated hydraulic press. Dense pellets so obtained were silver electrode by using a fine paintbrush to coat both faces of the pellets with a thin layer of silver paint. Thus, coated pellet was used for the electric measurements. Electrical properties were determined by impedance analyzer (Precision Component analyzer by Wayne Kerr Electronics). This instrument directly provides the values of capacitance (C) and dielectric loss ($\tan\delta$) which were used to calculate other electric quantities. The data was recorded at room temperature in the frequency range of 20 Hz–3 MHz in air. The magnetic data were recorded by using a Vibrating Sample Magnetometer (Lakeshore 665).

3. Results and discussion

3.1. Powder X-ray diffraction

The powder X-ray diffraction patterns of different $\text{Sr}_{1-x}\text{La}_x\text{Fe}_{12}\text{O}_{19}$ samples (where $x = 0, 0.08, 0.13, 0.18$) are shown in Fig. 1. It is

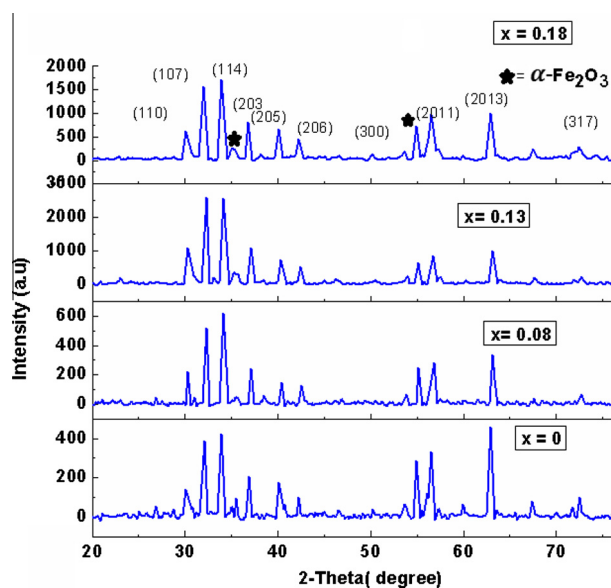


Fig. 1. XRD patterns of $\text{Sr}_{1-x}\text{La}_x\text{Fe}_{12}\text{O}_{19}$ ($x = 0, 0.08, 0.13, 0.18$).

observed that all the samples with lanthanum doping concentrations are composed of M-type hexagonal phase having space group P63/mmc. Also, the intensities and the position of diffraction peaks are at the same position as that of standard $\text{SrFe}_{12}\text{O}_{19}$ (JCPDS Card No. 00-043-0002), and matches well as reported by others [21,22]. Some traces of impurities like $\alpha\text{-Fe}_2\text{O}_3$ (JCPDS Card No. 15-615) were also detected (as shown in Fig. 1) indicating incomplete reaction between various reactants. It needs higher and longer sintering to remove these undesired peaks (impurities). However, the absence of any La_2O_3 peaks in the XRD patterns suggests that Sr^{2+} ions are substituted by La^{3+} ions in the structure [23]. In the structure of strontium hexaferrite, Fe^{3+} ions are distributed over five different sites: three octahedral sites (2a, 12k and 4f2), one tetrahedral site (4f1) and one bipyramidal site (2b). In the La-substituted structures, La^{3+} ions are expected to enter the Sr^{2+} sites because of their compatibility in radius; the radius of La^{3+} (1.22 Å) being comparable to that of Sr^{2+} (1.32 Å). The substitution of La at Sr site leads to a valence change of Fe^{3+} to Fe^{2+} at 2a or 4f2 site to conserve the charge neutrality [13]. Many investigators have reported the substitution of La at Sr sites in the lanthanum doped strontium hexaferrites [23,24].

The lattice parameters ('a' and 'c') and unit cell volume (V_{cell}) of all the three samples are calculated from the XRD data of the main peaks according to the following relations and the results are listed in Table 1.

$$\frac{1}{d^2} = \frac{4}{3} \left(\frac{h^2 + hk + k^2}{a^2} \right) + \frac{l^2}{c^2}; \quad V_{\text{cell}} = 0.8666a^2c$$

The lattice constant 'a' was found to be almost constant while 'c' continuously decreases with the increase of La^{3+} concentration and is in agreement with the literature [24,25]. The most probable reason of change in 'c' may be due to the fact the ionic radius of the La^{3+} (1.22 Å) is smaller than Sr^{2+} (1.32 Å) and that of Fe^{2+} (0.80 Å) is larger than Fe^{3+} (0.67 Å). So the replacement of Sr^{2+} by La^{3+} and change of Fe^{3+} to Fe^{2+} results in change of lattice parameters [26].

The average grain size of all the three samples were calculated from the main two peaks using the Scherrer's equation $D = \frac{0.89\lambda}{\beta \cos\theta}$, where λ is the incident wave length, β is the full width half maximum and θ is the angle considered. The values calculated are listed in Table 1. It was observed that the grain size decreases with the La concentration as reported earlier [23] and is attributed to the fact that La ion is entering into the ferrite lattice. In the present investigation, the grain size lies in the range of 29.71–76.49 nm and is smaller than those reported earlier [26].

3.2. Energy dispersive spectroscopy results

Energy Dispersive X-rays Spectroscopy (EDS) of the samples was carried out to confirm the formation of pure and doped strontium hexaferrite. Fig. 2(a) and (b) shows EDS spectra of pure and La doped strontium hexaferrite. The presence of strontium, lanthanum, iron and oxygen in the samples confirms the formation of the required material.

The SEM micrograph of the surface of a typical La doped strontium hexaferrite ($\text{Sr}_{0.82}\text{La}_{0.18}\text{Fe}_{12}\text{O}_{19}$) sample is shown in Fig. 3 at a

Table 1
Grain size, lattice parameters and unit cell volume of $\text{Sr}_{1-x}\text{La}_x\text{Fe}_{12}\text{O}_{19}$ samples.

Samples	Grain size (nm)	$a = b$ (Å)	c (Å)	c/a	V_{cell} (Å ³)
($\text{SrFe}_{12}\text{O}_{19}$)	76.49	5.882	23.23	3.949	696.49
($\text{Sr}_{0.92}\text{La}_{0.08}\text{Fe}_{12}\text{O}_{19}$)	33.28	5.884	23.05	3.917	691.56
($\text{Sr}_{0.87}\text{La}_{0.13}\text{Fe}_{12}\text{O}_{19}$)	31.22	5.886	22.98	3.904	689.93
$\text{Sr}_{0.82}\text{La}_{0.18}\text{Fe}_{12}\text{O}_{19}$	29.17	5.888	22.88	3.885	687.40

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