



# Electrically and magneto-dielectrically modified $\text{La}^{3+}$ doped $\text{Co}_{0.3}\text{Li}_{0.1}\text{Ca}_{0.5}\text{Fe}_{2.1-x}\text{La}_x\text{O}_4$ spinel ferrites and their potential applications

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## ARTICLE INFO

### Keywords:

Resistivity  
Tangent loss  
High-frequency  
Bolometers  
Crystal Structure

## ABSTRACT

Low temperature and more efficient sol-gel auto combustion process was utilized to synthesize  $\text{Co}_{0.3}\text{Li}_{0.1}\text{Ca}_{0.5}\text{Fe}_{2.1-x}\text{La}_x\text{O}_4$  [La-CLCF] spinel ferrites (where  $x = 0.00, 0.01, 0.02, 0.03, 0.04$ , and  $0.05$ ). The La-CLCF sample's structural, optoelectrical, dielectric, and magnetic properties were examined as a result of  $\text{La}^{3+}$  doping. All synthesized La-CLCF samples have single-phased spinel matrix, according to X-ray diffraction (XRD) analysis. It was found that the crystallite size was 11 nm, and the bandgap energy of 3.45 eV for  $\text{La}^{3+}$  doping  $x = 0.03$ . The resistivity and activation energy both were increased with the substitution of  $\text{La}^{3+}$  in the CLCF spinel matrix, while the peak position of temperature coefficient of resistance (TCR) percentage was  $-3.92\%/K$  at 372 K for sample  $x = 0.03$ . The alternating current conductivity improves with frequency, whereas the dielectric constant and tangent loss decrease as frequency increases. Further, the saturation magnetization and the microwave operating frequency for all the samples were reduced with the doping of  $\text{La}^{3+}$ . The minimum coercivity was 242 Oe at  $x = 0.03$ . Our study suggested that the  $\text{Co}_{0.3}\text{Li}_{0.1}\text{Ca}_{0.5}\text{Fe}_{2.07}\text{La}_{0.03}\text{O}_4$  sample is tested as an active material for potential use in high-frequency applications, electromagnetic wave absorbing material, and evaluating the sensitivity of infrared (IR) detectors used in night vision bolometers.

## 1. Introduction

Soft ferrites possess a low coercivity value, making them readily demagnetizable, and find widespread application in the electronics industry. They are utilized in various electronic components such as inductors, microwaves, and transformers, due to their high resistivity, permeability, temperature stability, cost-effectiveness, and minimal energy losses [1]. Moreover, the materials with higher temperature coefficient of resistance are useful for room-temperature infrared bolometers, and magnetic sensors [2–4]. Due to their low dielectric loss, excellent temperature performance, high resistivity, inexpensive, and high remanence ratio, lithium ( $\text{Li}^{1+}$ ) and doped lithium ( $\text{Li}^{1+}$ ) ferrites are interesting materials for microwave devices [5]. As laminating

ferrite layers for multilayer chip inductors (MLCIs),  $\text{Li}^{1+}$  ferrites are also appropriate due to their excellent electromagnetic properties at higher frequencies, low sintering temperatures, and high Curie temperature [6]. Lithium ferrite exhibits a spinel-type crystal structure with the general formula  $\text{AB}_2\text{O}_4$ . Within this structure, metal cations ( $\text{Li}^{1+}$  and  $\text{Fe}^{2+/3+}$ ) are distributed across both tetrahedral (A-) and octahedral (B-) sites. The arrangement of  $\text{Li}^{1+}$  and  $\text{Fe}^{2+/3+}$  ions is a critical factor influencing the properties of lithium ferrite. In the crystal lattice, a deficiency of lithium can result in cation defects, the formation of secondary phases, and shortages of lithium, which is known for its relatively volatile nature [7].

Many researchers worked on the  $\text{Li}^{1+}$  and  $\text{Li}^{1+}$  substituted spinel ferrites (SFs) including Li-Co [8], Li-Ca [9], and Li-La [10] spinel

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<https://doi.org/10.1016/j.jalcom.2023.173015>

Received 20 April 2023; Received in revised form 15 October 2023; Accepted 27 November 2023

Available online 30 November 2023

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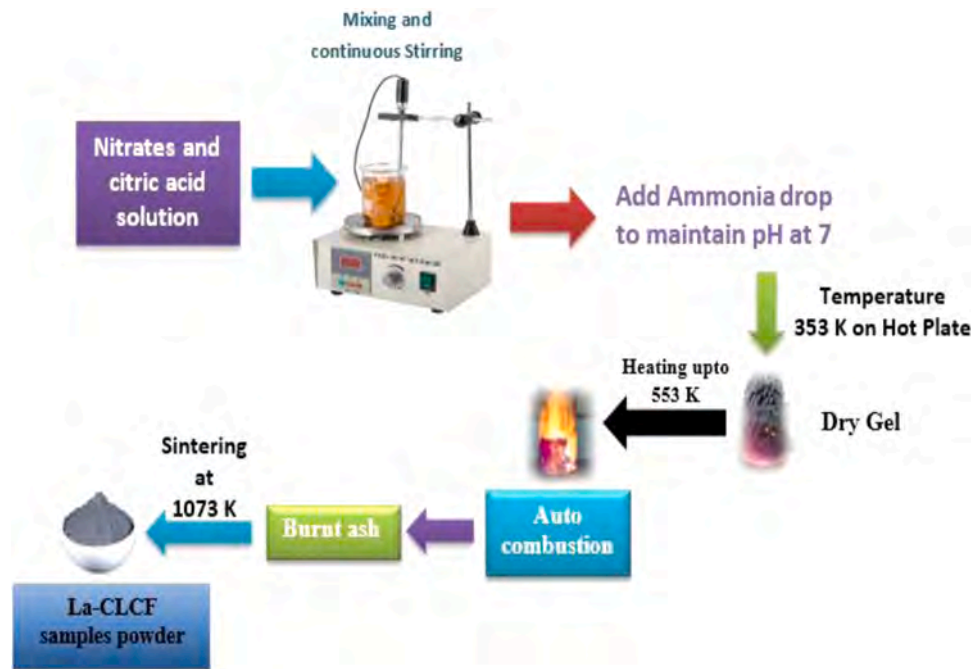


Fig. 1. SGAC process for the synthesis of La-CLCF samples.

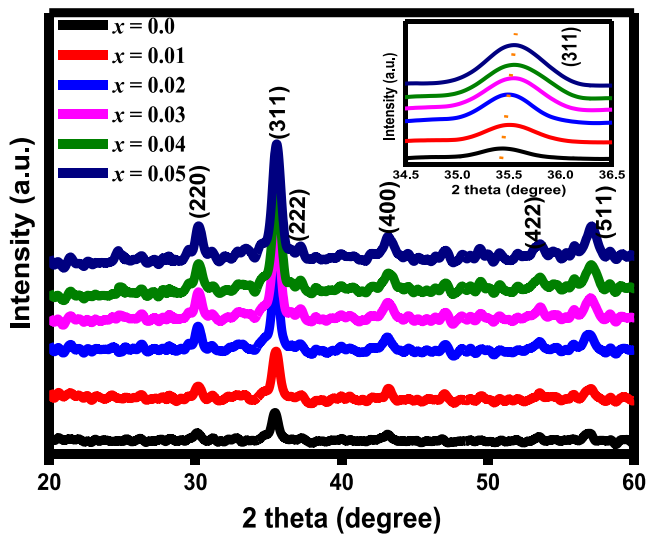


Fig. 2. XRD spectra for La-CLCF samples.

ferrites. For lithium-ion batteries (LIBs) anode, the non-toxic and inexpensive  $Li_{1.1}Co_{0.3}Fe_{2.1}O_4$  ferrite was demonstrated to be a superior electrochemical material in terms of rate capability, and specific capacity [8]. Randhawa *et al.*, synthesized  $Li_{0.25}Ca_{0.5}Fe_{2.25}O_4$  SFs using the solution combustion route and reported the crystallite size was 30 nm, whereas the lattice constant was 8.464 Å. At room temperature (RT), the tangent loss had small values, while the dielectric constant had high values suggesting it is useful for high-frequency devices [9]. Sattar *et al.*, [10] reported  $Li_{0.5}Fe_{2.5-x}La_xO_4$  ( $x = 0.0$  and 0.1) SFs fabricated via the ceramic route. They observed that the substitution of rare earth  $La^{3+}$  ion does not affect the  $Li^{1+}$  ferrite's structural and magnetic behavior. Therefore, the insertion of various dopant ions into the lithium ferrite lattice results in materials with novel and intriguing characteristics. The variations in these characteristics are caused by a cation distribution among the ferrite sublattice's tetrahedral (A-) and octahedral (B-) sites. This is due to the fact that the degree of inversion in doped  $Li^{1+}$  ferrites,

and thus their parameters, are observed to be highly dependent on the pH value, preparation route, sintering temperature and time, amount and type of the dopant [11].

The precise characteristics of SFs, both in terms of their physical and chemical properties, are intricately dependent on the synthesis method employed, as well as a range of other factors including temperature, pH, and the choice of raw materials [12,13]. The hydrothermal route [14], co-precipitation route [15–18], sol-gel auto-combustion (SGAC) process [19–23], a solid-state method [24], and other synthetic procedures have been used by different researchers. Because of its benefits, such as low cost, the capability to control the grain size, simple method, and easy setup [25], we have preferred the SGAC method to prepare ferrites. In the current study, the impact of doping  $La^{3+}$  concentration on  $Co_{0.3}Li_{0.1}Ca_{0.5}Fe_{2.1-x}La_xO_4$  [La-CLCF] where  $x = 0.00, 0.01, 0.02, 0.03, 0.04$ , and 0.05 SFs prepared using the SGAC process was examined. The main focus is to observe the impact of  $La^{3+}$  on structural, morphological, and optical bandgap energy, resistivity, dielectric, and magnetic behavior of La-CLCF spinel ferrites.

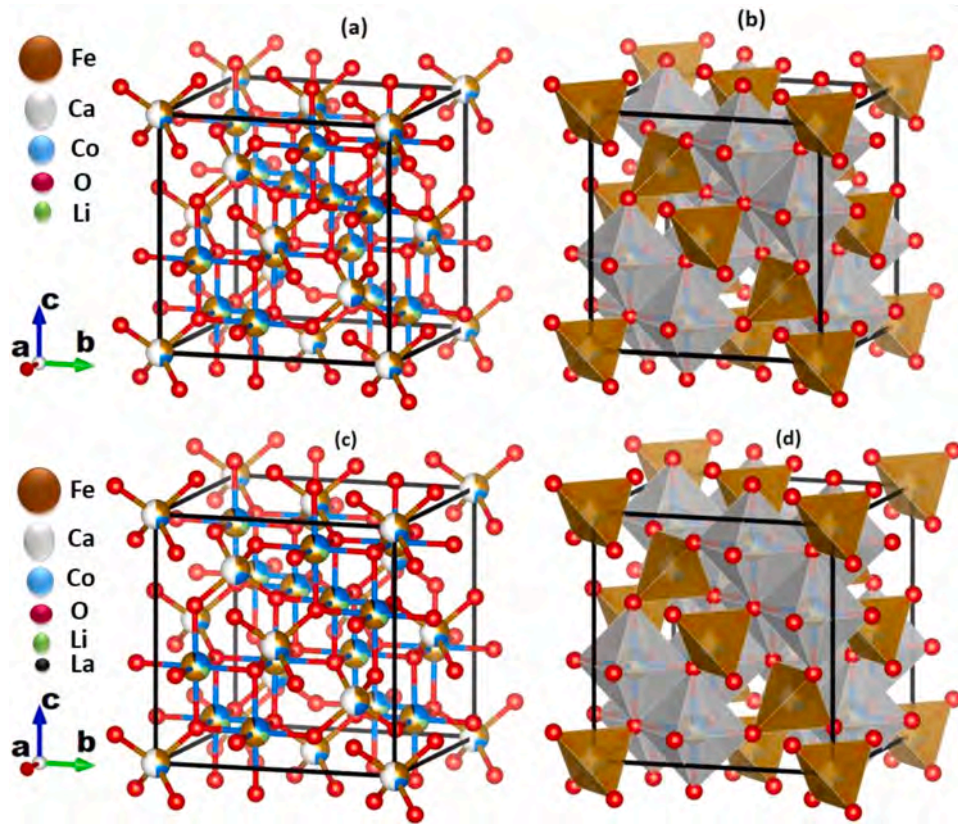
## 2. Experimental parts

### 2.1. Materials used

The metal nitrates including  $La(NO_3)_3 \cdot 6 H_2O$  (98.6%),  $Ca(NO_3)_2 \cdot 6 H_2O$  (97.9%),  $Co(NO_3)_2 \cdot 6 H_2O$  (97.9%),  $Fe(NO_3)_3 \cdot 9 H_2O$  (97%),  $LiNO_3 \cdot 6 H_2O$  (99.99%), citric acid ( $C_6H_8O_7$ ), and ammonia solution were utilized as starting materials to synthesize La-CLCF samples powder.

### 2.2. Method of preparation

The SGAC process was used to prepare the sample powder. In this process, first, the metal nitrates solution and citric acid were prepared in a beaker with a molar ratio of 1:1, according to the stoichiometric calculation in deionized water. The breaker is put on magnetic stirring and adds ammonia solution to maintain the pH~7. After achieving the required solution pH, the temperature was maintained at 353 K of the magnetic stirrer, and continuous stirring. The solution first transforms into a gel and then the temperature of the magnetic stirrer is up to 553 K.



**Fig. 3.** (a) Unit cell structure for CLCF sample (b) tetrahedral and octahedral sites in the unit cell structure of CLCF sample (c) unit cell structure for sample  $x = 0.01$  (d) tetrahedral and octahedral sites in unit cell structure for sample  $x = 0.01$ .

**Table 1**  
Structural parameters for La-CLCF samples.

$x$	$D$ (nm)	$d$ (Å)	$a$ (Å)	$\delta$ (nm <sup>-2</sup> )	$V$ (Å <sup>3</sup> )	$H_A$ (Å)	$H_B$ (Å)	$r_p$ (Å)	$d_x$ (g/cm <sup>3</sup> )	$\rho_B$ (g/cm <sup>3</sup> )	$\rho_R$ (%)	$P$ (%)	$d_x$ (g/cm <sup>3</sup> )
0.00	13	2.496	8.28	0.0051	567	3.585	2.927	0.741	5.140	3.01	170.78	41	5.140
0.01	15	2.500	8.29	0.0041	570	3.590	2.931	0.743	5.139	3.16	162.63	38	5.139
0.02	14	2.508	8.31	0.0050	575	3.602	2.940	0.745	5.108	3.39	150.70	33	5.108
0.03	11	2.515	8.34	0.0078	580	3.611	2.948	0.747	5.086	3.58	142.08	29	5.086
0.04	10	2.518	8.35	0.0083	582	3.616	2.952	0.748	5.084	3.85	132.07	24	5.084
0.05	10	2.529	8.38	0.0091	590	3.632	2.965	0.751	5.037	3.96	127.21	21	5.037

The formed gel was transferred into ash with auto ignition and performed sintering at 1073 K for 8 h. The fluffy loose powder after sintering was grounded to change into well-fine powder. The SGAC preparation route for La-CLCF samples is illustrated in Fig. 1.

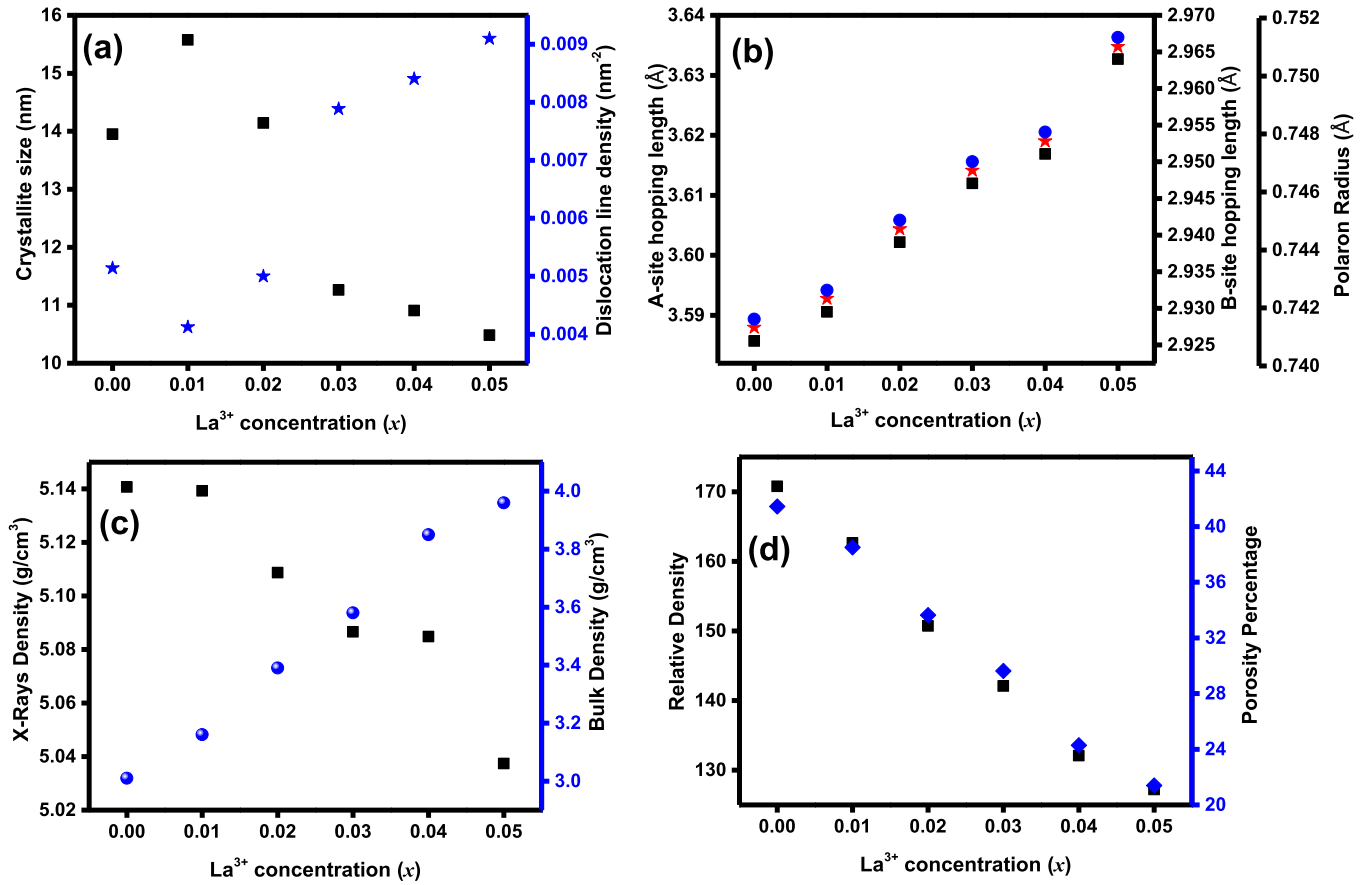
### 2.3. Characterization

To obtain a Bragg angle ( $2\theta$ ) in the range of  $20^\circ - 60^\circ$ , a D8 Bruker Advanced X-ray diffractometer having Cu-K $\alpha$  radiation with a wavelength ( $\lambda$ ) of 1.5418 Å was used. Scanning Electron Microscope (SEM), Nova Nano series 450 was utilized to record the micrographs of the La-CLCF material. UV-visible spectroscopy was performed to record the wavelength and absorbance to measure the energy bandgap using Perkin Elmer, Model Lambda 25, UV-visible double spectrometer (UV-DS). KEITHLEY, Model 2401 source meter, IV Measurement Meter utilized two probes technique to measure the temperature-dependent resistivity. IM3536 series LCR meter was utilized for recorded dielectric parameters, and a vibrating sample magnetometer (VSM), Model VSM-175 was employed to measure the hysteresis loop.

## 3. Results and discussion

### 3.1. Structural analysis

La-CLCF powder XRD spectra are depicted in Fig. 2 and the zoom part of the most intense (311) peak indicates a peak shift towards a greater angle with doping of La<sup>3+</sup> in CLCF samples. It was observed that six peaks at different  $2\theta$  angles correspond to (220), (311), (222), (400), (422), and (511) planes of the spinel matrix [19,26]. Fig. 3(a) indicates unit cell structure and Fig. 3(b) represents A- and B- sites in unit cell structure for the CLCF sample. Similarly, Fig. 3(c) indicates unit cell structure, and Fig. 3(d) represents A- and B- sites in unit cell structure for sample  $x = 0.01$ . The interplanar spacing ( $d$ ), experimental lattice constant ( $a$ ), and unit cell volume ( $V$ ) were investigated [26] and these parameters were increased with the replacement of Fe<sup>3+</sup> by La<sup>3+</sup> in La-CLCF samples (as seen in Table 1). The peak shift and change lattice parameters may be due to the replacement of smaller radii Fe<sup>3+</sup> (0.641 Å) with greater radii La<sup>3+</sup> (1.154 Å) ions [27]. The crystallite size ( $D$ ) was calculated [28,29], and it was observed that it was 13 nm for  $x = 0.0$  and decreased from 15 nm to 10 nm with the replacement of La<sup>3+</sup> ions (as reported in Table 1). It may be due to La<sup>3+</sup>-O<sup>2-</sup> having greater bond energy than Fe<sup>3+</sup>-O<sup>2-</sup>, therefore high energy is required to



**Fig. 4.**  $\text{La}^{3+}$  concentration ( $x$ ) versus (a) crystallite size and dislocation line density (b) A- and B- sites hopping length with polaron radius (c) X-ray and bulk densities (d) relative density and porosity for all the La-CLCF samples.



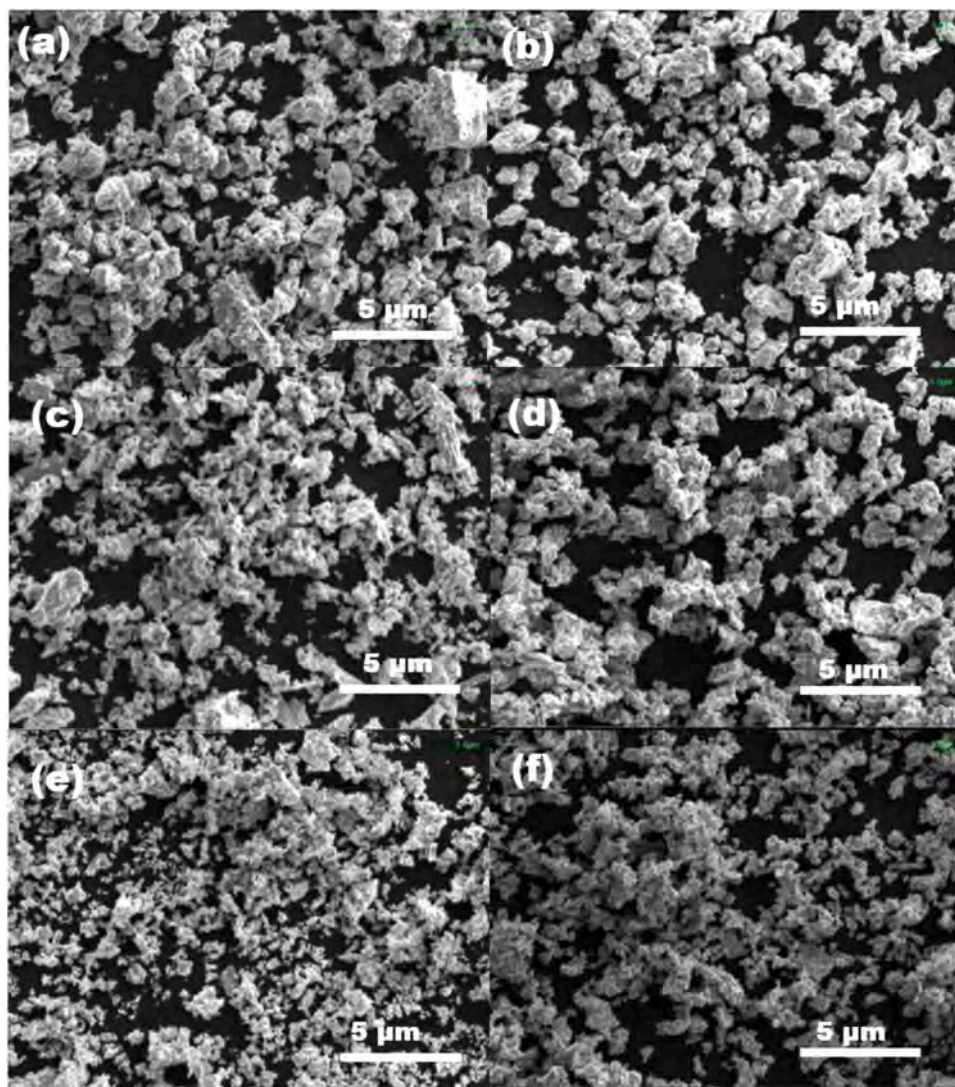


Fig. 5. SEM micrographs for La-CLCF samples (a) La = 0.0 (b) La = 0.01 (c) La = 0.02 (d) La = 0.03 (e) La = 0.04 (f) La = 0.05 at 5  $\mu\text{m}$  scale.

add  $\text{La}^{3+}$  ions into *B*-site. This limits the crystal growth and crystallization, resulting in reduced crystallite sizes with the doping of  $\text{La}^{3+}$  [30]. The dislocation line density ( $\delta$ ) was determined and the “ $\delta$ ” was increased with decreasing “*D*” and *vice versa* [28]. The calculated values are given in Table 1 and it was observed that “ $\delta$ ” increases with the doping of the  $\text{La}^{3+}$  ion in the CLCF spinel matrix. The graphical picture of “*D*” and “ $\delta$ ” *versus*  $\text{La}^{3+}$  concentration (*x*) is depicted in Fig. 4(a). The *A*-site hopping length ( $L_A$ ), *B*-site hopping length ( $L_B$ ), and polaron radius ( $\gamma$ ) were investigated [28], and the hopping length was linked with the lattice constant. Therefore, *A*- site hopping lengths, *B*- site hopping lengths, and polaron radius increase with the replacement of  $\text{La}^{3+}$  ions (as reported in Table 1). The  $L_A$ ,  $L_B$ , and  $\gamma$  all enhance with the doping of  $\text{La}^{3+}$  in the CLCF lattice (as seen in Fig. 4(b)). The X-ray ( $d_X$ ), bulk ( $d_B$ ) and relative ( $d_R$ ) densities were determined [28,31], and listed in Table 1. The X-ray and relative densities reduce and bulk density increase with the replacement of dopant ions. The graphical representation of  $d_X$ , and  $d_B$  densities with  $\text{La}^{3+}$  content (*x*) is depicted in Fig. 4 (c). The porosity percentage (*P*%) of as-prepared La-CLCF samples was investigated [31], and the porosity percentage reduced from 41% to 21% for *x* = 0.0 to *x* = 0.05 and the graphical picture of *P* (%) *versus*  $\text{La}^{3+}$  doping (*x*) and relative density is depicted in Fig. 4(d). The decrease in the porosity percentage may be due to the increase of bulk density [32] and this can also arise through the substitution of  $\text{Fe}^{3+}$  ions

with larger  $\text{La}^{3+}$  ions, resulting in reduced porosity [33].

### 3.2. Morphological analysis

SEM micrographs of La-CLCF power samples as depicted in Fig. 5(a-f). Because of the magnetic nature of SFs, all of the samples revealed a significant degree of agglomeration in irregular shape particle structure, and the particles are non-uniformly distributed. It can be seen from Fig. 5(a-f) that the agglomeration increases with the insertion of  $\text{La}^{3+}$  in the CLCF lattice.  $\text{La}^{3+}$  ions are larger compared to the host  $\text{Fe}^{3+}$  cations in the material. The size disparity can result in stronger electrostatic attraction particles. These attractive forces promote particle agglomeration as they tend to pull particles closer together [34].

### 3.3. Optical energy bandgap analysis

The bandgap energy ( $E_g$ ) of La-CLCF samples was calculated using the Tauc-Plots formula as depicted in Fig. 6(a-f). The Tauc relation is utilized to find the bandgap energy ( $E_g$ ) [35];

$$\alpha h\nu = B(h\nu - E_g)^n \quad (1)$$

The incident photon energy, transition probability constant, and absorption coefficient are all represented by  $h\nu$ , *A*, and  $\alpha$  respectively.

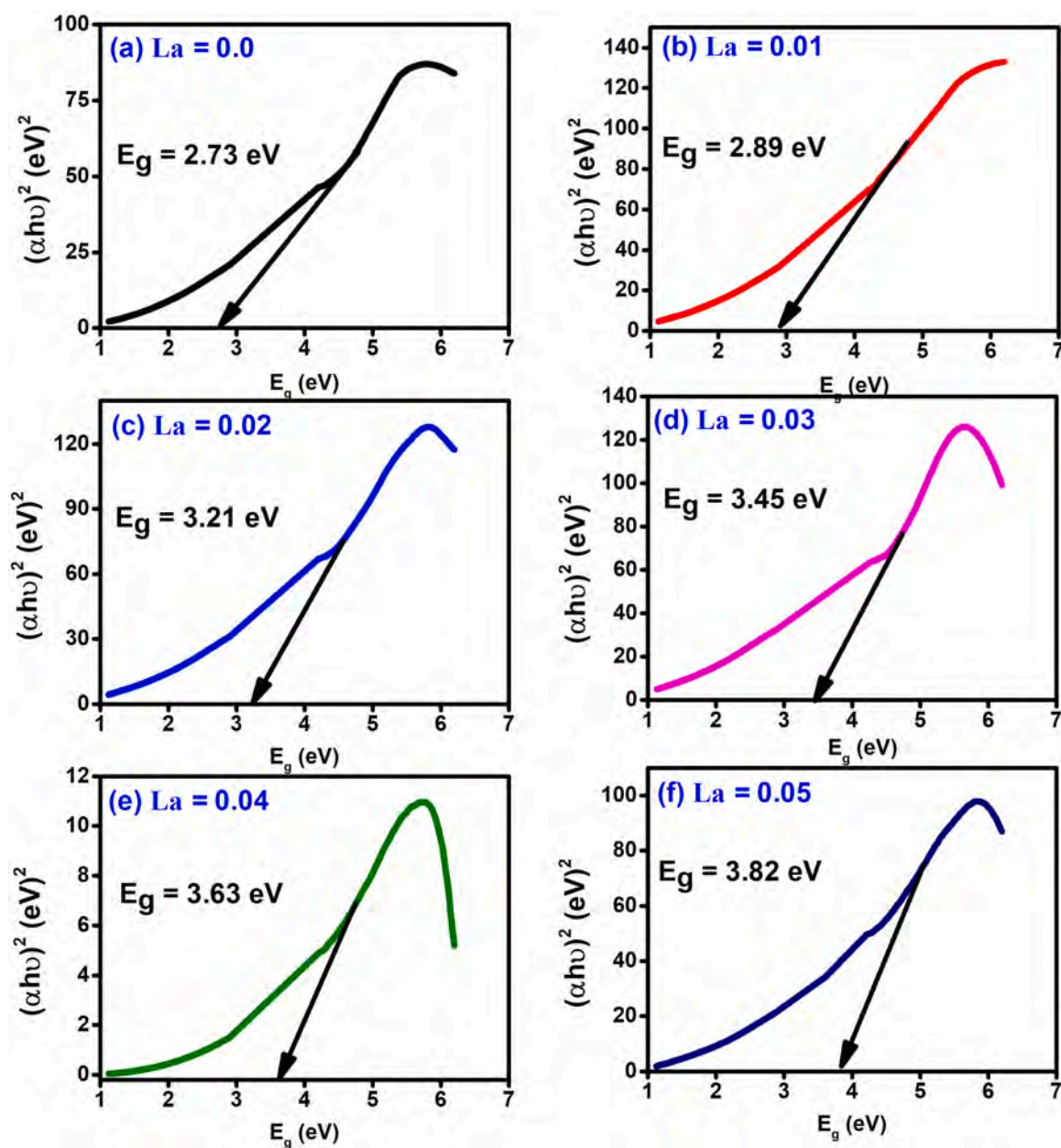


Fig. 6. Tauc plots for all the La-CLCF samples (a) La = 0.0 (b) La = 0.01 (c) La = 0.02 (d) La = 0.03 (e) La = 0.04 (f) La = 0.05.

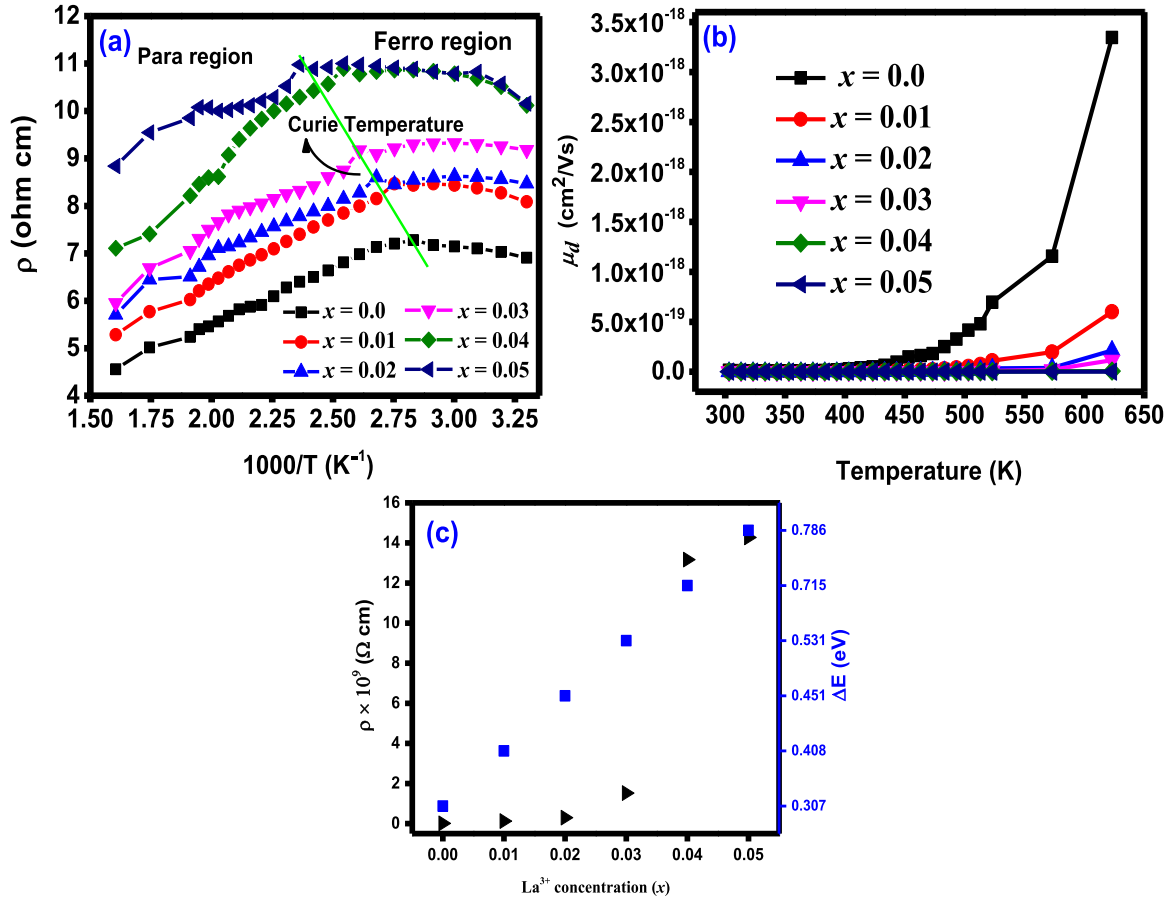


Fig. 7. (a) Arrhenius plots (b) drift mobility versus temperature for La-CLCF samples (c) resistivity at 303 K, and activation energy versus  $\text{La}^{3+}$  concentration.

**Table 2**  
Electrical parameters of La-CLCF samples.

$x$	$\rho \times 10^9$ at 303 K ( $\Omega$ cm)	$\eta \times 10^{22}$	$\mu_d \times 10^{-21}$ at 303 K ( $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ )	$\Delta E$ (eV)	$T_C$ (K)
0.00	0.008	1.732	14.8835	0.307	353
0.01	0.122	1.803	0.9491	0.408	363
0.02	0.295	1.918	0.3678	0.451	373
0.03	1.523	2.008	0.0681	0.531	383
0.04	13.167	2.141	0.0074	0.715	393
0.05	14.275	2.184	0.0068	0.786	403

Depending on whether the transition is direct or indirect, the constant 'n' equals 1/2 or 2. Fig. 5(a-f) shows the bandgap energies of La-CLCF samples. When the  $\text{La}^{3+}$  doping ( $x$ ) increases from 0.0 to 0.05, the value of " $E_g$ " increases from 2.73 to 3.82 eV. The incorporation of  $\text{La}^{3+}$  ions into the host lattice can cause changes in its crystal structure and lattice parameters. These structural modifications can affect the band structure, leading to an increase in the bandgap energy. The larger size of  $\text{La}^{3+}$  ions may introduce strain and alter the spacing between atoms, which in turn affects the electronic energy levels. The decrease in bandgap energy results in increasing crystallite size and vice versa [36]. Therefore, in our case with decreasing the crystallite size (as observed in XRD), the bandgap energy was increased.

### 3.4. Electrical analysis

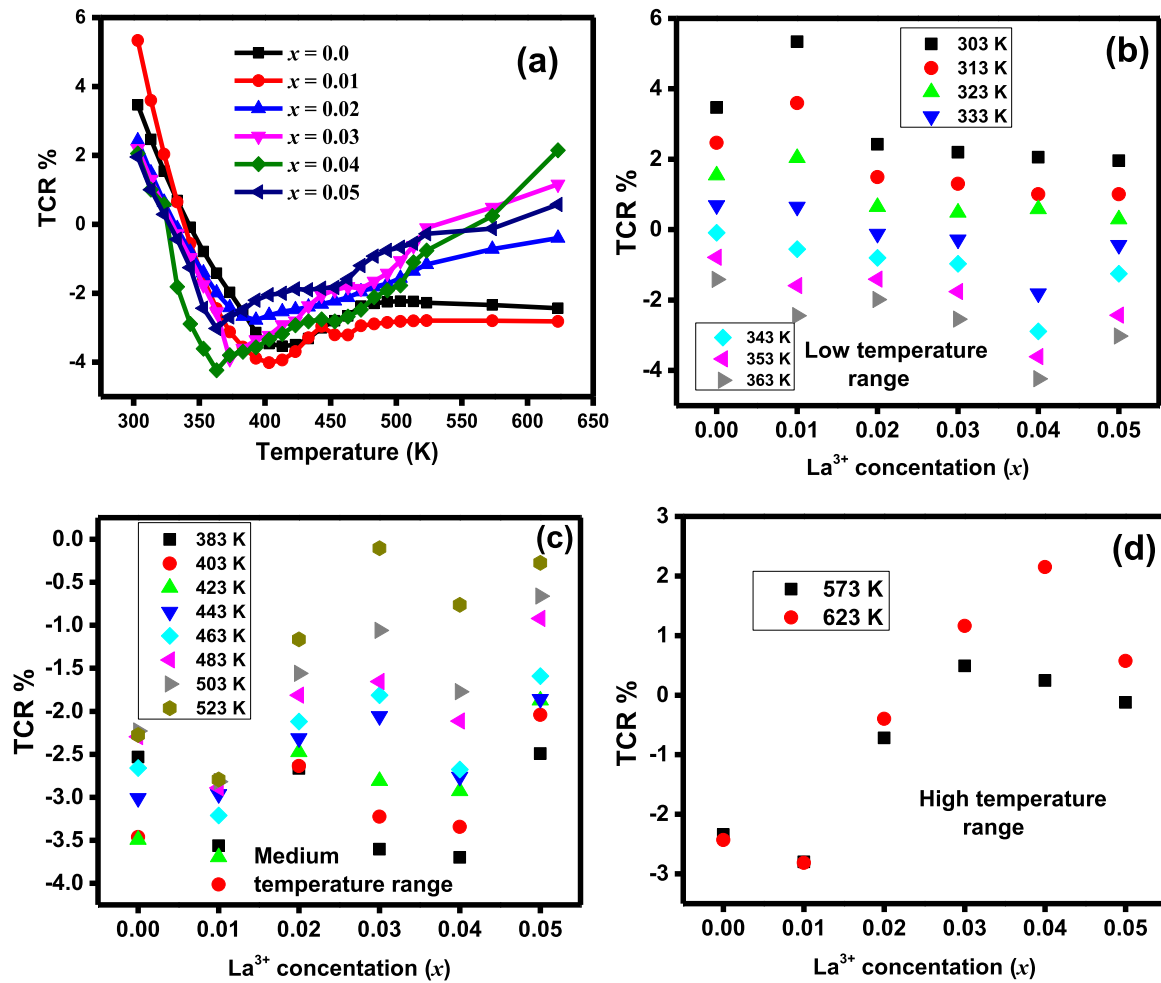
Typically the resistivity ( $\rho$ ) of SFs has ranged from  $10^5 \Omega$  cm to  $10^9 \Omega$  cm, and it can be influenced by some parameters, including the kind of dopant cation, the sintering temperature, and the method of preparation. In this instance, the relation  $\rho = RA/L$  is used to calculate the

resistivity, where  $L$  and  $A$  indicate the thickness and cross-sectional area of the pellets, respectively, while  $R$  stands for the resistance. According to the Arrhenius relation,  $\rho = \rho_0 e^{\Delta E/k_B T}$ , the resistivity decreases with increasing temperature, demonstrating the semiconductor nature of the SFs [37]. The resistivity ( $\rho$ ) versus  $1000/T$  (Arrhenius plots) of La-CLCF samples as seen in Fig. 7(a) are distributed into two linear regions. The first low-temperature region (Ferro region), and the resistivity are found to increase with increasing temperature. On the other hand, in the second high-temperature region (Para region), the resistivity decreases with increasing temperature. The exchange of electrons between the elements in more than one valence state and scattered unsystematically over lattice sites can be used to explain the conduction mechanism of ferrites [38]. Higher resistivity is the result of the  $\text{La}^{3+}$  ion replacements in the CLCF lattice. Every  $\text{La}^{3+}$  substitution may result in a reduction in the hopping between iron ions, such as  $\text{Fe}^{2+} \rightarrow \text{Fe}^{3+}$  at B- sites, which helps to marginally enhance the resistivity. From Table 2, it was clear that the resistivity at RT (303 K) was reduced from  $0.008 \times 10^9 \Omega$  cm to  $14.275 \times 10^9 \Omega$  cm (graphically as seen in Fig. 7(c)). Table 2 lists the Curie temperatures ( $T_C$ ) for various compositions of the La-CLCF samples as obtained from Arrhenius plots (as seen in Fig. 7(a)). In Fig. 7(a), it is evident that " $T_C$ " exhibits a trend to drop as  $\text{La}^{3+}$  content increases. The drift mobility ( $\mu_d$ ) was determined using the equation [39]:

$$\mu_d = \frac{1}{\eta e \rho} \quad (2)$$

where  $\eta = \frac{N_a d q P_{Fe}}{M}$  is the concentration of electric charge carriers ( $M$ ,  $P_{Fe}$  and  $N_a$  indicates molecular weight, number of Fe atoms, and Avogadro number) and  $e = 4.8032451 \times 10^{-10}$  esu (charge of an electron). Higher resistivity SFs are found to have low drift mobility, and vice versa. The drift mobility versus temperature is depicted in Fig. 7(b). Fig. 7(b) shows





**Fig. 8.** (a) Temperature coefficient of resistance *versus* temperature plots (b) Temperature coefficient of resistance *versus* La<sup>3+</sup> concentration (x) at temperature 303–363 K (c) Temperature coefficient of resistance *versus* La<sup>3+</sup> concentration (x) at temperature 383–523 K (d) temperature coefficient of resistance *versus* La<sup>3+</sup> concentration (x) at temperature 573–623 K.

that when the temperature rises, mobility rises as well. According to Table 2, a rise in La<sup>3+</sup> contents causes a decrease in drift mobility ( $\mu_d$ ) of the charge carriers but not the electric carrier concentration ( $\eta$ ). This shows that rather than a change in carrier concentration, the variation in resistivity with temperature in the mixed La-CLCF ferrite system is caused by a change in charge carrier drift mobility. The activation energy ( $\Delta E$ ) in SFs originates from the motion of the charge carrier. The hopping process and activation energy are closely related to one another, and the resistivity follows the same trend. The “ $\Delta E$ ” is also affected by the substitution of La<sup>3+</sup> ions and determined using relation (3) which is given in Table 2.

$$\Delta E = 2.303 \times k_B \times 1000 \times \text{slope}(eV) \quad (3)$$

The “ $\Delta E$ ” was increased from 0.30 eV to 0.78 eV as the La<sup>3+</sup> doping increased from  $x = 0.0$  to  $x = 0.06$  (graphically as seen in Fig. 7(c)). For evaluating the sensitivity of infrared detectors (IR) used in night vision bolometers, and magnetic sensors, the temperature coefficient of resistance (TCR) is extremely valuable [2–4,40,41]. The TCR data for pure and La-doped CLCF samples *versus* temperature were examined and illustrated in Fig. 8(a). The values of TCR% obtained from the peak position of Fig. 8(a) for La-CLCF samples are  $-3.54\%/K$  at 413 K ( $x = 0.0$ ),  $-4.04\%/K$  at 402 K ( $x = 0.01$ ),  $-2.77\%/K$  at 393 K ( $x = 0.02$ ),  $-3.92\%/K$  at 372 K ( $x = 0.03$ ),  $-4.21\%/K$  at 363 K ( $x = 0.04$ ), and  $-3.03\%/K$  at 362 K ( $x = 0.05$ ). Fig. 8(b) shows La<sup>3+</sup> concentration (x) *versus* TCR% at temperatures 303–363 K (low-temperature range) and it was found the TCR% value maximum at

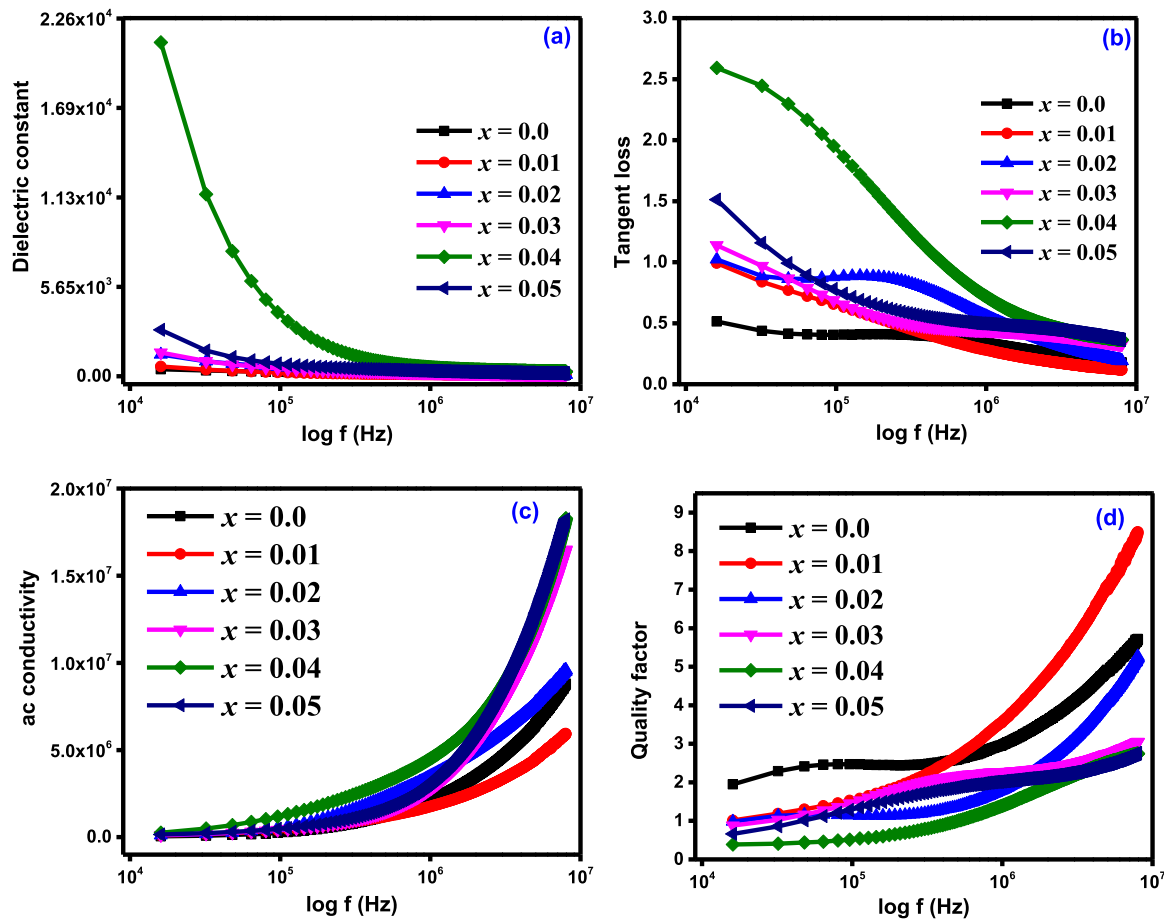
$x = 0.01$ . Fig. 8(c) shows La<sup>3+</sup> concentration (x) *versus* TCR% at temperatures 383–523 K (medium temperature range) and it was found the TCR% value was maximum at  $x = 0.03$ . Finally, in Fig. 8(d) La<sup>3+</sup> contents (x) *versus* TCR% at temperatures 573–623 K (high-temperature range) is shown, and the TCR% value was maximum at  $x = 0.04$ .

### 3.5. Dielectric analysis

The La-CLCF samples dielectric constant ( $\epsilon'$ ), dielectric tangent loss ( $\tan \delta$ ), ac conductivity ( $\sigma_{ac}$ ) and quality factor (Q factor) *versus* the log of frequency ( $\log f$ ) were measured. Fig. 9(a) showed the  $\epsilon'$  *versus* the log  $f$  revealing a high  $\epsilon'$  at low frequencies and a small value at high frequencies. The  $\epsilon'$  decreases with frequency as a result of the electric dipoles' inability to align with the alternating electric field's rapid change. The Maxwell-Wagner model can explain interfacial polarization, a frequent occurrence in materials that are consistent with the well-known Koop's hypothesis [42,43]. Charges that are restricted at grain boundaries enter the grains at high frequencies, lowering grain resistance and resulting in a low value of  $\epsilon'$ . At low frequencies, the  $\epsilon'$  of the samples was observed to be unexpectedly high.

Tangent loss ( $\tan$ ) is the term for the energy lost by the dielectric material when the dipoles are aligned to the electric field and lesser values of “ $\tan \delta$ ” indicate more practical application [44]. The frequency response of  $\tan \delta$  for the La-CLCF samples is depicted in Fig. 9(b). The loss, which is particularly significant at around  $10^2$  kHz, reduced





**Fig. 9.** (a) Dielectric constant versus the log of frequency for La-CLCF samples (b) Tangent loss versus the log of frequency for La-CLCF samples (c) ac conductivity versus the log of frequency for La-CLCF samples (d) Quality factor versus the log of frequency for La-CLCF samples.

dramatically with rising frequency, eventually becoming frequency-independent at a higher frequency. In a dielectric material, the “ $\tan \delta$ ” value governs the amount of energy dissipated. Koop’s theory, which is based on polarization resonance with the applied field, can explain this behavior. According to Koop’s theory, grain boundaries have higher resistance at lower frequencies, therefore hopping electrons from one site to another requires more energy. As a result, at lower frequencies, energy loss is high. In contrast, grain boundaries have lower resistivity at higher frequencies, requiring less energy for electrons to move between two ions at lattice sites [45].

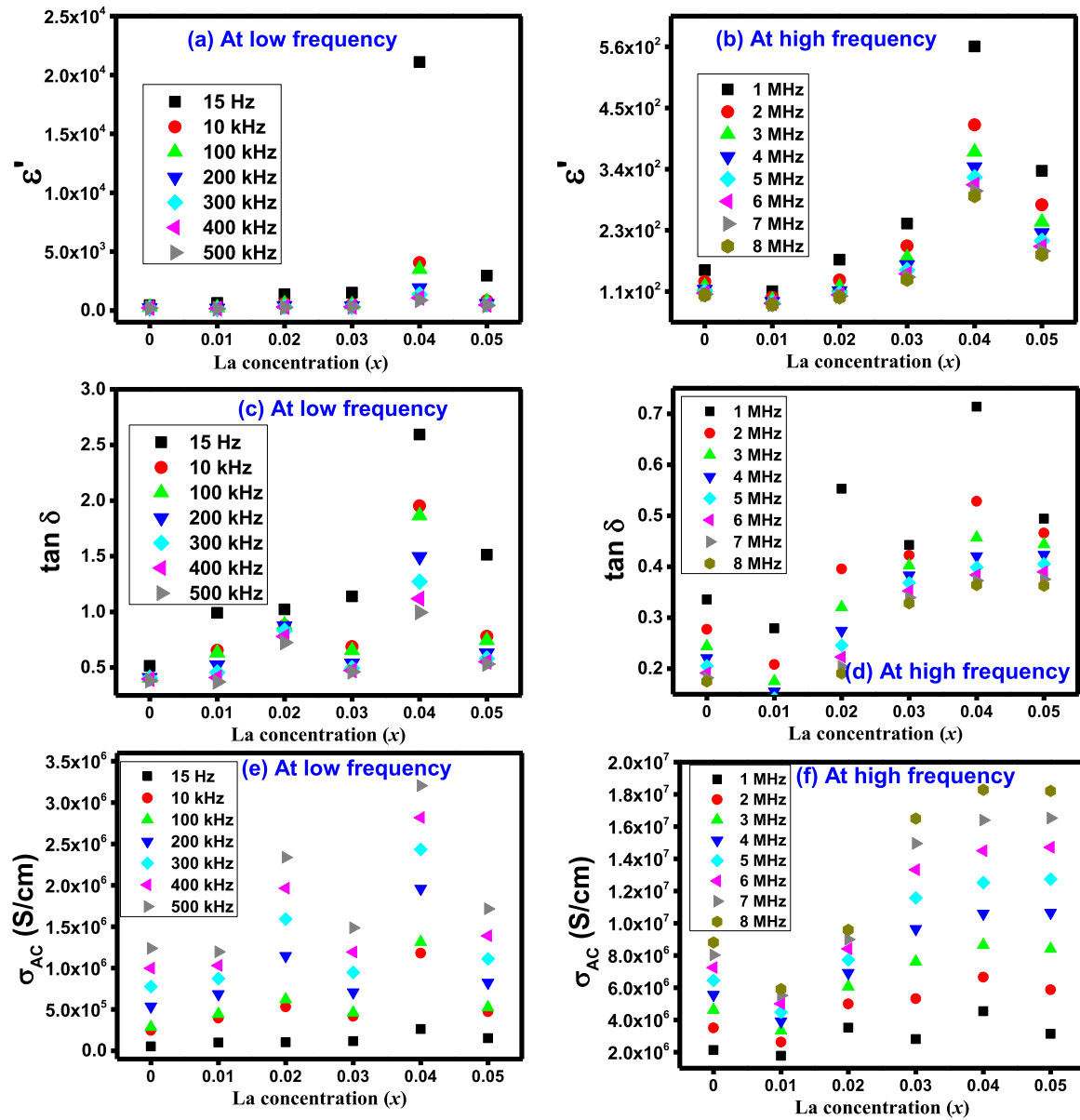
The frequency response of the La-CLCF samples ac conductivity ( $\sigma_{ac}$ ) is shown in Fig. 9(c). For all of the samples, the “ $\sigma_{ac}$ ” was excessively low at lower frequencies and the hopping of charge carriers between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  lattice sites were responsible for the increase in “ $\sigma_{ac}$ ” at high frequencies. Maxwell–Wagner’s and Koop’s theories can also explain the variation in ac conductivity. According to these theories, the dielectric structure is composed of two layers: The grain boundaries between well-conducting ferrite grains are formed by a thin layer of poorly conducting material. At low frequencies, poor-conducting grain boundaries become more active, leading to small ac conductivity, whereas high-conductivity ferrite grains are effective at high frequencies, resulting in larger ac conductivity and decreased electron hopping frequency between the ions.

To estimate the efficiency of some electrical devices, the quality factor (Q-factor) examines the link between stored energy and the rate of energy dissipation. The Q-factor of ferrite materials is an important factor in the design of electrical devices. The variations in the Q-factor of La-CLCF samples with frequency are depicted in Fig. 9(d). It was clear from Fig. 9(d) that the Q-factor increased with the applied frequency.

Fig. 10(a) showed that  $\text{La}^{3+}$  concentration ( $x$ ) versus dielectric constant at frequencies 15 Hz to 500 kHz (low-frequency range) and the dielectric constant was found to be maximum at  $x = 0.04$ . In Fig. 10(b) of  $\text{La}^{3+}$  concentration ( $x$ ) versus dielectric constant at high frequency 1–8 MHz, the dielectric constant was seen also as a maximum at  $x = 0.04$ . Fig. 10(c) showed that  $\text{La}^{3+}$  doping ( $x$ ) versus “ $\tan \delta$ ” at low frequencies 15 Hz to 500 kHz and the “ $\tan \delta$ ” minimum at  $x = 0.0$ , while, in Fig. 10(d) of  $\text{La}^{3+}$  concentration ( $x$ ) versus tangent loss at high frequency 1–8 MHz, the tangent loss was observed minimum at  $x = 0.01$ . In Fig. 10(e) of  $\text{La}^{3+}$  concentration ( $x$ ) versus ac conductivity at frequency 15 Hz to 500 kHz (low-frequency range), the ac conductivity was minimum at  $x = 0.03$ , while Fig. 10(f) of that  $\text{La}^{3+}$  concentration ( $x$ ) versus ac conductivity at frequency 1–8 MHz (high-frequency range) revealed that the tangent loss was also minimum at  $x = 0.01$ .

### 3.6. Hysteresis loop analysis

Hysteresis loops ( $M$ – $H$  loops) of  $\text{La}^{3+}$  substituted CLCF samples with a low coercive field, are shown in Fig. 11(a). Table 3 revealed the values of all the magnetic parameters. The La-CLCF sample’s saturation magnetization ( $M_s$ ) and remanent magnetization ( $M_r$ ) were maximum for  $x = 0.01$ , while remanent magnetization was minimum (21 emu/g) at  $x = 0.03$  which may be due to nonmagnetic  $\text{La}^{3+}$  ions replacement in the CLCF spinel lattice. Moreover, the “ $D$ ” was reduced with the replacement of  $\text{La}^{3+}$  ions in the CLCF sample (as seen in Table 1). The introduction of  $\text{La}^{3+}$  content, with  $x = 0.01$ , results in an increase in  $M_s$  to 117 emu/g. Following this point, there is a gradual decrease in  $M_s$ , dropping to 83 emu/g with increasing  $\text{La}^{3+}$  doping. The magnetic characteristics of the SFs are substantially influenced by factors such as



**Fig. 10.** (a) Dielectric constant *versus* La<sup>3+</sup> concentration ( $x$ ) at low-frequency range (b) dielectric constant *versus* La<sup>3+</sup> concentration ( $x$ ) at high-frequency range (c) tangent loss *versus* La<sup>3+</sup> concentration ( $x$ ) at low-frequency range (d) tangent loss *versus* La<sup>3+</sup> concentration ( $x$ ) at high-frequency range (e) ac conductivity *versus* La<sup>3+</sup> concentration ( $x$ ) at low-frequency range (f) ac conductivity *versus* La<sup>3+</sup> concentration ( $x$ ) at high-frequency range.

cation distribution, particle size, and A-B interactions [46–49]. The notable increase in  $M_S$  for  $x = 0.01$  can be attributed to the redistribution of cations within the A- and B- sites. When non-magnetic La<sup>3+</sup> was substituted for Fe<sup>3+</sup> ( $5u_B$ ), which was the main contributor to the magnetic properties at B- site of the SFs, the result may be explained by the weak A-O-B super-exchange interaction. It is well known that the magnetic properties of SFs with the AB<sub>2</sub>O<sub>4</sub> spinel crystal matrix depend on the cation content at the A- and B- sites. AB<sub>2</sub>O<sub>4</sub> is characterized by exchange interactions (A-A) and (B-B), while superexchange interaction is represented by (A-B). In comparison to the (A-A) and (B-B) interactions, the (A-B) interaction exhibits significantly stronger interactions. Because of the occupancy of La<sup>3+</sup> ions, the concentration of Fe<sup>3+</sup> ions at the B- site of the SFs decreased, reducing the (B-B) interactions and, as a result, the (A-B) super-exchange interactions. In other words, the strongly magnetic Fe<sup>3+</sup> ( $5u_B$ ) ions are the main cause of the B- site magnetic behavior in SFs. As non-magnetic La<sup>3+</sup> ions gradually replaced magnetic Fe<sup>3+</sup> ions, the B-B (Fe<sup>3+</sup>-Fe<sup>3+</sup>) interactions decreased. Thus, a decrease in the magnetic moment with higher La<sup>3+</sup>

doping can be attributed to a reduction in the net magnetic moment [50, 51]. For  $x = 0.01$ , the maximum coercivity ( $H_C$ ) was found, and the minimum coercivity was 242 Oe for sample  $x = 0.03$ . The low coercivity indicates the soft nature of the materials and is useful for electromagnetic wave absorbing materials in C-band frequencies [52]. The coercivity depends on extrinsic factors such as grain size, surface effects, and the movement of the domain walls. Finally, the change in magnetic parameters may be due to the doping of the La<sup>3+</sup> ion with the Fe<sup>3+</sup> ion, which modifies the orientation of the Fe<sup>3+</sup> ion's magnetic moment, and the arrangement of Fe<sup>3+</sup> ions in the crystal structure is changed. The soft magnetic nature of La-CLCF SFs changed with La<sup>3+</sup> doping and is good for high-frequency applications, including circulators, and inductor cores. With increasing La<sup>3+</sup> content, the squareness ratio ( $S_q = Mr/Ms$ ) was decreased as reported in Table 3 and it was observed that the minimum  $S_q$  was 0.24 for  $x = 0.03$ . The value of " $S_q$ ", less than 0.5 represents particle magnetostatic interaction [43]. Therefore, the  $S_q$  ratio for all La-CLCF samples showed the interaction of particles by magnetostatic interaction. The anisotropy constant ( $K$ ) was determined

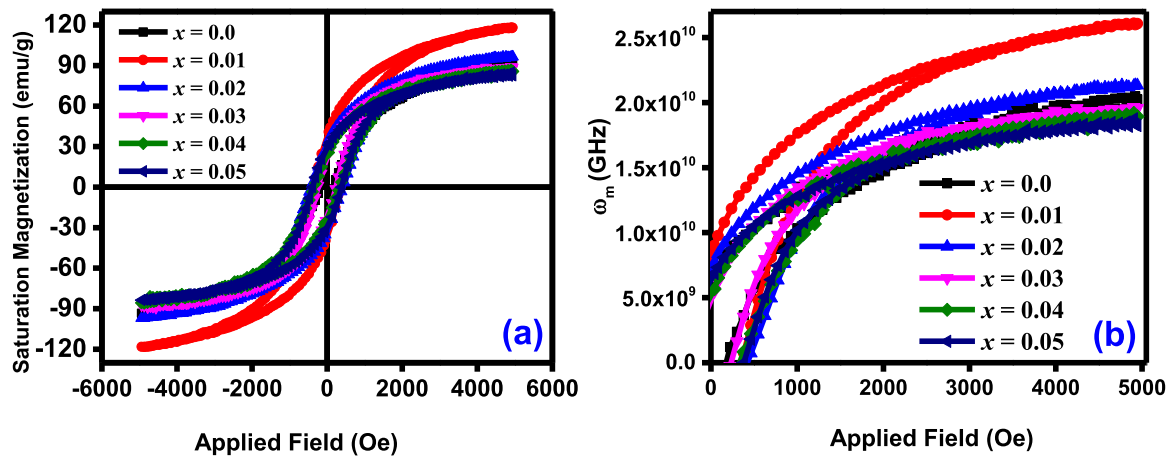


Fig. 11. (a) Hysteresis loops for La-CLCF samples (b) applied field versus microwave operating frequency for La-CLCF samples.

Table 3

Magnetic properties of La-CLCF samples.

x	$M_s$ (emu/g)	$M_r$ (emu/g)	$H_C$ (Oe)	$S_q$ ratio	$K$ (erg/cm <sup>3</sup> )	$\omega_m$ (GHz)
0.00	92	24	189	0.26	18,202	20.39
0.01	117	37	359	0.31	43,941	25.93
0.02	95	32	430	0.33	42,957	21.15
0.03	87	21	242	0.24	22,247	19.43
0.04	85	23	346	0.27	30,956	18.97
0.05	83	28	391	0.34	33,947	18.39

via the equation:  $K = \frac{H_C \times M_s}{0.9}$  [53] and the value of “K” (as seen in Table 3) is affected by  $M_s$  and  $H_C$ . The microwave operating frequency was calculated using the formula:  $\omega_m = 8\pi^2 M_s \gamma$  (where  $\gamma = 2.8$  MHz/Oe is the gyromagnetic ratio) [54–57] and values of “ $\omega_m$ ” was found to be in the gigahertz range as seen in Table 3. According to Fig. 11(b), La-CLCF samples are a favorable choice for high-frequency devices because of their microwave frequency response.

#### 4. Conclusions

In conclusion, the SGAC technique was used to successfully synthesize La-CLCF samples. Based on the XRD data, single-phase spinel-type ferrites were found in all the compositions. The energy bandgap plots showed a decreasing trend and had a value of 3.45 eV for sample  $x = 0.03$ . The resistivity was  $1.523 \times 10^9 \Omega \text{ cm}$  and the activation energy of 0.531 eV at  $x = 0.03$ , while the peak position of TCR was  $-3.92\%/K$  at 372 K for  $x = 0.03$ . When frequency increased, both the loss tangent and the dielectric constant experienced a sharp decline. The  $M_s$  was reduced with the insertion of  $\text{La}^{3+}$  and had a value of 87 emu/g, while small coercivity was 242 Oe observed for the sample  $x = 0.03$ . Due to its easy to fabricate, low cost, low dielectric tangent loss, high resistivity, small coercivity, and high TCR%, the current research revealed that the sample  $x = 0.03$  is effective for high-frequency applications, electromagnetic wave absorbing material, and evaluating the sensitivity of infrared detectors (IR) used in night vision bolometers.

#### Funds for fellowship

Muhammad Imran Arshad would like to thank HEC Pakistan for giving opportunity of Postdoc under post doc batch 3 (Ref: 3-1/PDFP/HEC/2022(B-3)/2320/02). He also extends his appreciation to University College London, London, U.K., for hosting this fellowship.

#### CRedit authorship contribution statement

**Muhammad Imran Arshad:** Supervision, Project administration. **Afeefa Dastgiri:** Writing – original draft. **Faisal Alresheedi:** Formal analysis. **Nasir Amin:** Validation. **Le Duc Tung:** Review. **Mongi Amami:** Resources. **Nguyen Thi Kim Thanh:** Data analysis and critical review. **Hira Zahid:** Data curation, Investigation. **Atta Ur Rehman:** Methodology, writing, Editing. **Safa Ezzine:** Resources, Formal analysis. **Jolly Jacob:** Review, Formal analysis.

#### Declaration of Competing Interest

The authors declare that there are no competing interests to declare among authors of this work.

#### Data availability

Data will be made available on request.

#### Acknowledgements

The authors extend their appreciation to the Deanship of Scientific Research at King Khalid University, Abha, Saudi Arabia, for funding this work through the Research Groups Program, under grant no. R.G.P.2. 233/44.

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