

Conduction mechanisms, relaxation dynamics, and magnetic properties of Se-substituted Sr nanohexaferrites

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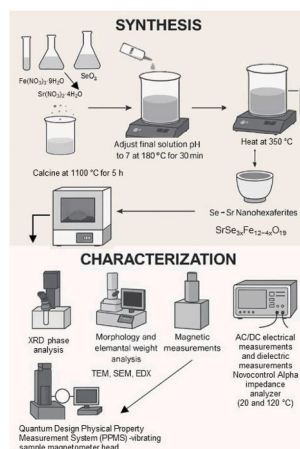
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HIGHLIGHTS

- Se-substituted Sr hexaferrites have been synthesized via sol-gel auto-combustion route.
- The magnetic, electrical, and dielectric features of NHFs have been studied.
- M_s , M_r and H_c increased as the Se concentration increased up to $x = 0.05$.
- Se substitution significantly influences the charge transport processes.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, Se-substituted Sr nanohexaferrites ($\text{SrSe}_{3x}\text{Fe}_{12-4x}\text{O}_{19}$ (Se→Sr) ($x \leq 0.10$) NHFs) have been synthesized via sol-gel combustion route. The X-ray diffraction technique was utilized in order to assess the phase formation of the samples. Furthermore, morphology and crystallinity were shown by SEM, TEM, and HR-TEM investigations. $M-H$ curves are consistent with the response of ferrimagnetic materials under an externally applied magnetic field. A “wasp-waist” behavior was also seen in the registered $M-H$ hysteresis loops. The magnetic investigations displayed an increased tendency for M_s , M_r and H_c as the concentration of selenium (Se) ions increased, achieved their maximum values for the sample with $x = 0.05$, then reduced as the concentration

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of Se ions further increased ($x > 0.05$). The maximum values achieved for the product with $x = 0.05$ might be useful in permanent magnet applications. This study also investigates the electrical and dielectric properties of Se-substituted Sr-NHFs through AC and DC conductivity measurements, activation energy analysis, and impedance spectroscopy. Results show that Se substitution modifies charge transport by enhancing hopping and thermally activated conduction mechanisms. AC conductivity displays power-law frequency dependence, while DC conductivity reveals temperature-activated behavior. Dielectric measurements indicate reduced dielectric constant and loss at higher frequencies due to interfacial polarization. Cole–Cole and Nyquist analyses confirm non-Debye relaxation and highlight changes in grain and grain boundary effects with Se content. These findings demonstrate the material's potential for high-frequency devices, energy storage, and electromagnetic shielding applications.

1. Introduction

M-type Hexaferrites (M-HFs) having the chemical formula of $\text{MFe}_{12}\text{O}_{19}$ ($\text{M} = \text{Sr}, \text{Ba}, \text{Pb}$), are classified as hard magnets due to the super-exchange interactions between Fe^{3+} ions at the different sublattices and blocks. Due to their high saturation magnetization, high chemical and thermal stability, low electric conductivity and eddy currents, high ferromagnetic Curie temperature, and low cost of production, they have a potential in energy storage devices such as super-capacitor, magnetic hoses for the transportation of magnetic fields, audio devices like speakers microwave and electronic devices for mobile technology, millimeter devices and electromagnetic wave absorbers, and refrigerator magnets, etc. [1]. They are magnetic semiconductors with a high value of bandgap, resistivity of $\sim 10^9$ Ohm.cm order, and stable at room temperature (RT). Their magnetic, electrical, optical, and dielectric properties depend on the preparation conditions (synthesis and calcination temperature, soaking time, and the type of doped ions). Besides high spontaneous magnetic moment and high resistivity at RT, they have high values of permittivity and permeability, low electric conductivity and eddy currents, resistance to corrosion, and high Curie temperature ($T_C = 472$ °C) [2–5]. Among the various M-type hexaferrites, strontium hexaferrite ($\text{SrFe}_{12}\text{O}_{19}$) is preferred over other M-type hexaferrites due to its exceptional properties, such as high saturation magnetization, high Curie temperature, and a large coercivity due to its high magneto-crystalline anisotropy [6]. The intrinsic dielectric properties of M-HFs are connected to their unfamiliar structure, specifically, the trigonal bipyramid. The mobility of electrons among the Fe^{2+} and Fe^{3+} ions, situated at the Oh site, is the main reason for the electrical conduction behavior of the HFs, which causes low resistivity as hopping of electrons will occur extremely easily hence conduction [7–9]. The intrinsic magnetic properties of these hexaferrites are controlled and modified by substitution of foreign ions [10]. The substitutions of cations in M-type hexaferrites have improved materials properties, and results show that substitution can improve their structural, magnetic, and electrical properties [11]. Patel et al. [11] investigated the influence of Co^{4+} - Ca^{2+} substitution on structural, microstructure, magnetic, electrical, and impedance characteristics of M-type barium–strontium hexagonal ferrites. Chauhan et al. [12] studied structural, magnetic, and dielectric properties of Co–Zr substituted M-type calcium hexagonal ferrite nanoparticles.

This is the first study in which Se→Sr NHFs have been synthesized via the sol-gel route and their structural, morphological, magnetic, and electrical/dielectric features have been investigated in detail.

2. Experimental

Se→Sr NHFs were synthesized through sol-gel auto-combustion by using $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (97 %, Merck), $\text{Sr}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ (98 %, Merck), SeO_2 (98 %, Sigma Aldrich). Firstly, a specific amount of metal nitrates, Se precursor and citric acid were thawed in deionized water (DI) and the temperature was set at 80 °C under stirring for 20 min. Ammonium solution was used to adjust the final solution pH to 7 and settled at 180 °C for 30 min, then the temperature was raised to 350 °C to transform the whole solution to gel, then burned to form a black powder.

Lastly, the powder was calcined at 1100 °C for 5 h to form the hexagonal ferrite as shown in Scheme 1 (Supplementary Materials).

Rigaku Miniflex 600 with $\text{CuK}\alpha$ was used for the phase analysis of the products. The morphology and elemental weight% of the products were analyzed by TEM and SEM along with EDX. The magnetic measurements were performed using a Quantum Design Physical Property Measurement System (PPMS) coupled with a vibrating sample magnetometer head. The proton conductivity studies and dielectric measurements of the samples were performed using a Novocontrol Alpha impedance analyzer (20 and 120 °C). The films were sandwiched between gold blocking electrodes, and the conductivities were measured in the frequency range 0.1 Hz–3 MHz at 10 °C intervals.

3. Results & discussion

3.1. Structure and morphology

XRD analysis of Se→Sr NHFs offers significant insights into the structural behavior of hexaferrites in the presence of Se doping. The hexaferrite phase is confirmed by the diffraction peaks that were seen in the patterns and the Sr hexaferrite unit cell [13], as shown in Fig. 1(a and b) corresponding to the single-phase of $\text{SrFe}_{12}\text{O}_{19}$ (PDF-2 00–079–1411). The small peak observed between (107) and (114) could be attributed to a secondary phase within the prepared samples. The X-ray density (ρ_x), the lattice parameters (a , c), the c/a ratio, the unit cell volume (V), the average crystal size, R_{Bragg} , refined quality factors R_{wp} , R_p , χ^2 and atomic positions were all estimated by the application of refinement using Match 4 and Fullproof software and listed in Table 1 and Table S2. The influence of Se doping on the lattice parameters (a and c) is presented in Table 1. It illustrates the variance that occurred as a result of the distortion that was brought about by the distribution of cations across the crystal. The Scherrer equation was used to estimate the size of the crystal, and when the (107) peak was taken into consideration, the measurements were found to fall within the range of 62–97 nm. The c/a ratio for all Se→Sr NHFs was calculated and found to be in the range of 3.91. According to the findings of the current study, the " c/a " ratio is lower than 3.98, which indicates that the materials that were synthesized have a structure that is of the M-type hexagonal ferrite variety.

Fig. 2 displays the SEM images of Se→Sr ($x = 0.00, 0.02, 0.04$, and 0.08) NHFs. The images depict agglomerated and irregular particle morphologies. The shape of the particles and the degree of agglomeration have a significant impact on the rising Se content. It was found that the particle size was smaller than 100 nm. Fig. 3 illustrates the elemental analysis, mapping, and weight and atomic percentages of Se→Sr NHFs ($x = 0.03$ and 0.05) NHFs, which are determined via EDX. The EDX spectra indicated the presence of elements such as Sr, Se, Fe, and O, which aligns with the XRD phase analysis of Se-doped Sr hexaferrite. The TEM and HR-TEM micrographs of Se→Sr NHFs ($x = 0.05$) NHFs are displayed in Fig. 4. An aggregation of hexagonal particles was visible in the TEM. GATAN software was used to calculate the values of interplanar spacing and the crystal planes from the HR-TEM image. The interplanar spacing values were found to be 0.10, 0.14, 0.20, and 0.21 nm, which equate to (414), (312), (207), and (206).

3.2. Magnetic properties

It became widely known that it is feasible to manipulate the electrical and magnetic features of M-type hexaferrites by varying the magnetic connections of iron Fe–O–Fe ion bonds and purposefully replacing them (or some other ions) in specific crystallographic locations with paramagnetic and/or diamagnetic ions. For instance, doping M-type hexaferrite material with diamagnetic and paramagnetic ions might result in frustrating the magnetism structure and decreasing its fundamental magnetic properties. Furthermore, the intra-lattice exchange interactions might be attenuated, resulting in a rise in the fundamental magnetic features of the M-type hexaferrites at the macro-sized level because of the lower compensating contributions of the superposition of the magnetic moments of two sublattices exhibiting ferrimagnetic features. Hence, the analysis of the magnetization curves of non-metal Se-doped M-type hexaferrite $\text{SrFe}_{12}\text{O}_{19}$ is of great interest.

Fig. 5 illustrates the results of investigating the magnetization versus magnetic field for Se→Sr NHFs measured at RT ($T = 300$ K) and low temperature ($T = 20$ K) and for magnetic fields $H = \pm 70$ kOe. The investigation of M–H curves at RT indicated that the different prepared Se→Sr NHFs reached saturation in magnetism at magnetic fields up to 70 kOe, as shown in Fig. 5(b) and (e). The hysteresis nature in the collected M–H curves is obvious, which is consistent with the response of ferrimagnetic materials under an externally applied magnetic field. The enlarged views of registered M–H plots around $H = 0$ kOe for different prepared materials, as shown in Fig. 5(c) and (f), indicated that the hysteresis loops are “wasp-waisted”. Such behavior suggests a coercive force distribution, which might be induced by the coexistence of various magnetic phases, the existence of superparamagnetic fractions, the bimodal grains’ distribution, or the dipolar interactions among

single-domain nano-sized particles [14]. Similar behavior was seen in the M–H loops of CoFe_2O_4 nano-sized particles, which display a reorientation of spins at low temperatures because of the spins’ canting effect on the nanoparticles’ surfaces [15]. Mironovich et al. [16] reported the influence of the ratios of Fe/Sr and Fe/Ba on the properties of M-type hexaferrite materials produced via the hydrothermal technique. The existence of SrFeO_{3-x} and BaFe_2O_4 resulted in wasp-waisted M–H loops, indicating that these two phases do not enter into interaction with hexaferrite, while samples of the products containing Fe_2O_3 have symmetrical M–H loops with no kinks, indicating good interactions among hexaferrite and hematite phases. In another study, Chithra et al. [17] revealed that the kinks observed in the M–H hysteresis loops of CoFe_2O_4 nanoparticles were generated by two different kinds of nanoparticles with distinct anisotropies. The plausible reasons for such behavior could be as follows: (i) the formation of secondary phases as revealed in the XRD patterns, (ii) the existence of a broad distribution of nano-sized particles with distinct anisotropies, and most probably (iii) the coexistence of single-domain and multi-domain grains at the same time.

Fig. 5(b) and (e) illustrate the field-dependence of magnetization at higher magnetic fields for better determination and analysis of the progress in the magnetic parameter, namely saturation magnetization (M_s). Additionally, Fig. 5(c) and (f) illustrate the field dependence of magnetization under a magnetic field ranging between -10 and $+10$ kOe for better determination and analysis of the progress in the magnetic parameters, namely M_r (remnant magnetization) and H_c (coercivity). According to these figures, an alteration in the quantities of M_s , M_r , and H_c is obviously produced by the rise in the amount of Se ions. The specific values of M_s were extracted from M–H hysteresis loops and presented in Fig. 6(a) and the evolution in M_r values is presented with

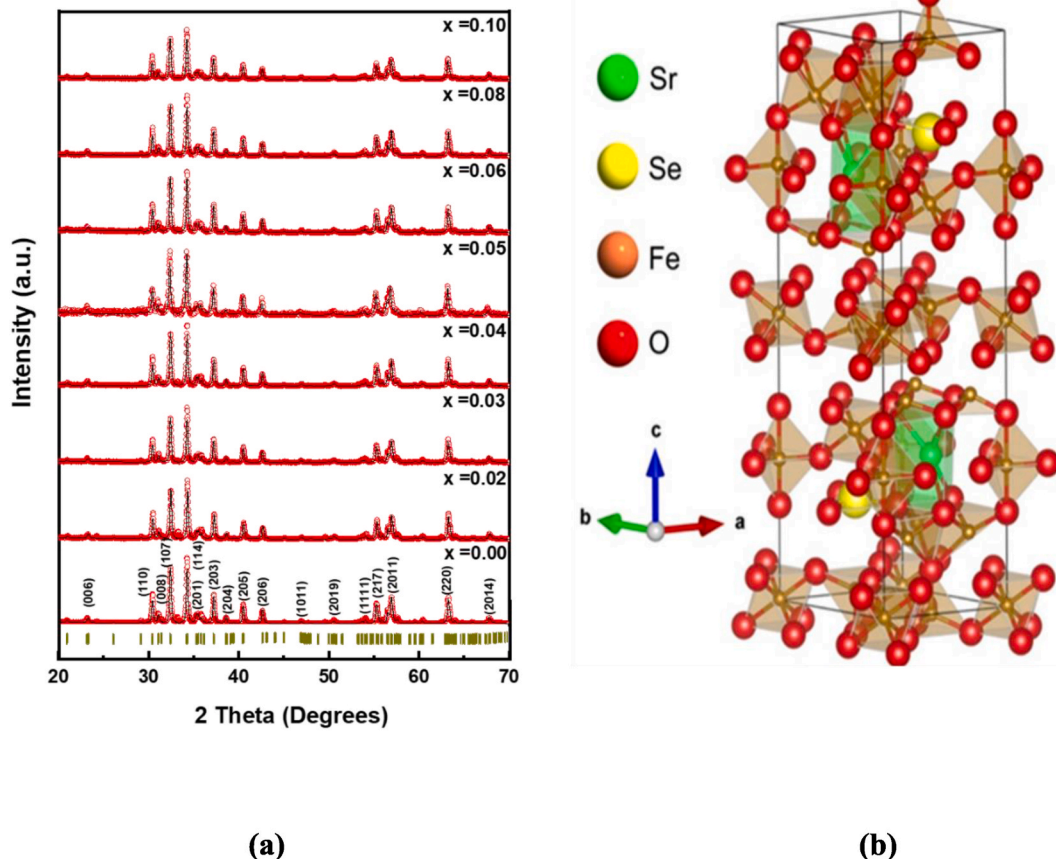


Fig. 1. (a) XRD powder patterns and (b) unit cell description of Se→Sr NHFs.

the Se ions concentration dependencies in Fig. 6(b). Compared to non-doped sample ($x = 0.00$), both M_s and M_r displayed an increased tendency as the concentration of Se ions rose up to $x = 0.05$, thereafter, reduced as the concentration of Se ions further increased. Indeed, M_s increased from 54.68 to 76.06 emu/g (approximately 39.1 % increase percentage) at 300 K and from 80.03 to 107.00 emu/g (approximately 33.7 % increase percentage) at 20 K with the rise in the Se content from $x = 0.00$ to 0.05. Thereafter, M_s reduced from 76.06 to 63.22 emu/g (approximately 16.9 % decrease percentage) at 300 K and from 107.00 to 92.67 emu/g (approximately 13.4 % decrease percentage) at 20 K with the rise in the Se content from $x = 0.05$ to 0.10. M_r increased from 26.89 to 37.33 emu/g (approximately 38.8 % increase percentage) at 300 K and from 39.73 to 53.68 emu/g (approximately 35.1 % increase percentage) at 20 K with the rise in the Se content from $x = 0.00$ to 0.05. Thereafter, M_r reduced from 37.33 to 30.85 emu/g (approximately 17.3 % decrease percentage) at 300 K and from 53.68 to 45.53 emu/g (approximately 15.2 % decrease percentage) at 20 K with the rise in the Se content from $x = 0.05$ to 0.10.

The variations in the magnetic saturation and remanence of Ba–Sr M-type hexaferrites are primarily determined by the following causes: (i) the residence of Fe ions at various sublattices of the hexaferrite cell, which has a direct connection to the strength of the exchange interactions, and (ii) the distortion of the magnetic structure at the surface of nano-sized particles that forms magnetically dead surface layers, which is known as spins' canting. (iii) The changes in the size of grains could also imply a surface compression of the crystal lattice. This might substantially raise the band gap. Nevertheless, bulk transport, particularly the transport of oxygen vacancies, will destroy the grains. In general, the exchange interactions typically align the magnetic moments of grains in a similar direction and subsequently enhance the magnetization. The M-type hexaferrite unit cell is typically composed of about twenty-four Fe^{3+} ions that reside in five distinct crystallographic positions: one bipyramidal sublattice (2b), one tetrahedral (Td) sublattice ($4f_1$), and three octahedral (Oh) sublattices (2a, $4f_2$, and 12k) [18]. For a magnetically ordered state, the $4f_1$ and $4f_2$ sublattices display spins in a downward direction ($\downarrow$), while those of 2a, 2b, and 12k are in the opposite direction ($\uparrow$). Considering the hypothesis that Se ions replaced the iron ions, the incorporation of paramagnetic Se ions into the sites with downward spins' direction would provoke a rise in the net magnetic moment of the hexaferrite system, and hence a boost in the saturation magnetization. Whereas the incorporation of paramagnetic Se ions into the sites with upward spins' direction will lead to a diminution in the net magnetic moment of the hexaferrite system, which in turn diminishes the saturation magnetization. Taking now into consideration the assumption that Se ions may replace Ba or Sr ions, the replacement of some diamagnetic ions (Ba^{2+} or Sr^{2+}) by paramagnetic Se ions would provoke a slight increase in the net magnetic moment of the M-type hexaferrite system, causing a marginal increase in saturation magnetization. However, if Se does not enter the crystal lattice and does not interact with the ions constituting the M-type hexaferrite system, a secondary phase will appear, which will evidently cause a reduction in saturation magnetization. The monotonic drop in M_s and M_r values at higher Se $x > 0.05$ may suggest that the paramagnetic substitution process disrupted the magnetism structure of Sr/Ba M-type hexaferrite

by breaking the long-range order of the exchange interactions. The addition of Se ions within the Sr–Ba M-type hexaferrite will disturb the lattice symmetry, leading strains within the M-type hexaferrite matrix, which in turn reduce the strength of Fe–O–Fe interactions, provoking a decline in the magnitude of magnetization. Besides the contribution of the disruption of the magnetic structure, which is the major reason for the reduction in M_s and M_r at higher Se content, the appearance of secondary phases, as shown from phase analysis data, might be an additional source for the drop in M_s and M_r . It is also obvious that the Se substitution, the magnetic moment (n_B):

$$n_B = (M_s \times MW) / 5585 \quad (1)$$

where MW is the molecular weight [19] (Equation (1)) of the prepared samples (Fig. 6(d)) increased with the increase in Se content and achieved its maximum for sample with $x = 0.05$, and then decreased with the further increase in the Se content ($x > 0.05$), which may explain the variations in M_s .

The square factor of the M – H loops, known as the squareness ratio $\text{SQR} = M_r/M_s$, is an important feature for determining the multi- and single-magnetic domain nature of a product and assessing its magnetic hardness. $\text{SQR} \leq 0.5$ is usually assigned to grains with multi-magnetic domains, while $\text{SQR} > 0.5$ is usually assigned to single-domain grains [20]. The domain rotation is the main driver in single-domain systems. In structures with arbitrarily oriented multi-domains, the force needed for orientation is small due to the fact that triggering the motion of domain walls needs a lesser amount of energy than activating domain rotation. Consequently, particles with single magnetic domains would exhibit strong coercivity. However, randomly oriented multi-magnetic domain particles would display smaller coercivity. As shown in Fig. 6 (e), the SQR values at both 300 and 20 K are more or less around 0.5 for different samples. In addition, there are some indications that multi-domain and single-domain grains can co-exist. Hence, the wasp-waisted M – H loops and the large values of coercive fields for different prepared compositions suggest that particles consisted of both single-magnetic domains and multi-magnetic domains.

The specific values of H_c were also extracted from M – H hysteresis loops and presented with the Se ions concentration dependencies in Fig. 6(f). As can be seen from this figure, compared to non-doped sample ($x = 0.00$), H_c at both 300 and 20 K displayed almost an increased tendency as the concentration of Se ions rose and achieved its maximum values of 6258.2 Oe (at 300K) and 5546.2 Oe (at 20K) for $x = 0.05$, then, reduced as the concentration of Se ions further increased ($x > 0.05$). For the present case, H_c increased from 4660.9 to 6258.2 Oe (approximately 34.3 % increase percentage) at 300 K and from 4529.7 to 5546.2 Oe (approximately 22.4 % increase percentage) at 20 K with the rise in the Se content from $x = 0.00$ to 0.05. Thereafter, H_c reduced from 6258.2 to 5018.1 Oe (approximately 19.8 % decrease percentage) at 300 K and from 5546.2 to 4594.4 Oe (approximately 17.1 % decrease percentage) at 20 K with the rise in the Se content from $x = 0.00$ to 0.10. The pre-dominant factors that alter the coercive field are the variation in grains size and the evolution in magneto-crystalline anisotropy [21]. As shown in Fig. 6(c), the magneto-crystalline anisotropy (K_{eff}) increased as the concentration of Se ions rose up to $x = 0.05$, then, reduced as the

Table 1
Structural parameters of Se→Sr NHFes.

x	a = b (Å)	c (Å)	c/a	D_{XRD} (nm)	$dx(\text{g}/\text{cm}^3)$	V (Å ³)	R_{Bragg}	R_p	R_{wp}	χ^2
0.00	5.8811	23.0459	3.91	71.8	4.974	690.3	14.1	21.2	27.6	2.5
0.02	5.8809	23.0401	3.91	79.6	4.976	690.1	14.5	22.0	28.4	2.6
0.03	5.8947	23.0403	3.91	97.3	4.975	690.2	13.9	21.2	26.8	2.2
0.04	5.8950	23.0945	3.91	62.2	4.940	695.0	13.6	23.3	28.5	2.6
0.05	5.9020	23.1671	3.92	77.9	4.947	698.8	20.8	35.7	43.2	1.2
0.06	5.8951	23.0963	3.91	88.6	4.981	695.1	13.0	21.5	25.1	1.7
0.08	5.8948	23.0927	3.91	72.6	4.941	694.9	11.0	20.3	25.7	2.0
0.10	5.8962	23.0981	3.91	89.8	4.972	695.4	11.3	19.8	24.5	1.6

concentration of Se ions further increased ($x > 0.05$). Such a tendency may explain the observed tendency in H_c values.

3.3. AC conductivity

The AC conductivity of Se→Sr NHFs increases with rising frequency, indicating strong frequency dependence. Conductivity varies slightly across different Se substitution ratios, ranging from $x = 0.01$ to 0.10 , with smaller intervals of 0.01 and 0.02 for the last two substitution levels. At low frequencies, conductivity increases with temperature, with this temperature dependence being particularly pronounced at $x = 0.06$ as shown in Fig. 7. The observed increase in AC conductivity with frequency suggests that the conduction mechanism in Se-substituted Sr-NHFs is predominantly frequency-dependent. This behavior aligns with mechanisms such as small polaron hopping or localized charge carrier dynamics, where the applied AC field facilitates charge carrier hopping between localized states. At higher frequencies, charge carriers respond more effectively to the oscillating field, resulting in enhanced conductivity. Such frequency-dependent behavior is well-documented in ferrite systems and is often attributed to Maxwell-Wagner polarization and conduction within grain boundaries [22–24].

The slight variation in AC conductivity across different Se substitution ratios ($x = 0.01$ to 0.10) suggests that Se incorporation modifies the electronic structure without significantly altering the conduction pathways. When Se ions substitute for oxygen or occupy interstitial sites, they may introduce localized states or influence defect density within the bandgap, affecting charge carrier hopping probabilities. However, these changes do not cause substantial variations in overall AC conductivity. The minimal difference observed at substitution intervals of 0.01 and 0.02 indicates that structural and electronic modifications induced by Se occur incrementally, potentially due to limited solubility or substitution saturation at higher concentrations. Similar trends have

been reported in other ferrite systems, such as Ti- and Zn-substituted ferrites, where substituents introduce localized changes in conduction without fundamentally altering the conduction mechanism [25–28].

The increase in AC conductivity at low frequencies with rising temperature highlights the role of thermally activated conduction processes, such as hopping of thermally excited carriers. Se-ion substitution influences this behavior by modifying the E_a required for charge carrier movement, likely through changes in grain boundary potential barriers or defect states. The extent of this influence varies with the substitution ratio, as Se incorporation may change grain boundary density or dielectric properties, both of which impact low-frequency conductivity. Reported studies suggest that the interaction between Se-induced defect states and thermally activated hopping mechanisms provides a plausible explanation for the observed behavior [29]. In ferrite materials, the combined effect of temperature-enhanced carrier mobility and low-frequency field-induced interfacial polarization is a well-documented phenomenon. The pronounced frequency dependence of AC conductivity observed here is consistent with the Jonscher universal power law, commonly reported in ferrite systems, where conductivity follows the relation (Equation (2)) [30,31]:

$$\sigma(\omega) = \sigma_0 + A\omega^n \quad (2)$$

where the exponent n offers valuable insight into the dominant hopping mechanism. Moreover, the incremental effect of Se substitution is consistent with trends in other ferrite systems, where substituent ions typically influence conductivity through subtle modifications to microstructure and defect chemistry, rather than inducing fundamental changes in the conduction mechanism. Similar behavior has been reported in ferrites, where increasing temperature activates charge carriers within grain boundaries and enhances hopping conduction processes [24,32]. The interplay between temperature and substitution

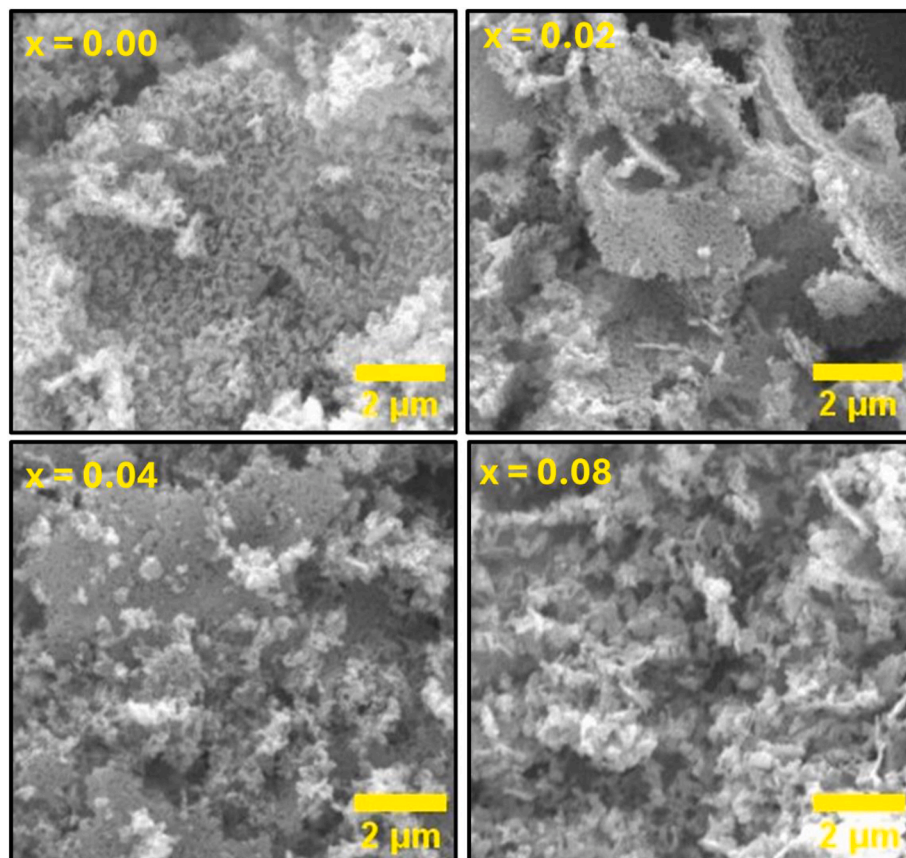


Fig. 2. SEM images of Se→Sr ($x = 0.00, 0.02, 0.04$, and 0.08) NHFs.

ratio is therefore particularly important for tailoring the material properties for targeted applications.

The AC conductivity of Se-substituted Sr-NHFs exhibits strong frequency dependence, slight variation with substitution ratios, and significant temperature effects at low frequencies, as depicted in Fig. 7. These behaviors are consistent with established models of polaron hopping and interfacial polarization in ferrite systems. Se substitution introduces localized electronic modifications that influence the material's overall dielectric and conductive properties. Further investigations, including impedance spectroscopy and microstructural analysis, would provide deeper insights into the underlying conduction mechanisms and the role of Se substitution in modifying material properties.

To evaluate the presence of polaron hopping in Se-substituted Sr-NHFs, AC conductivity measurements were analyzed using the Jonscher universal power law to determine whether the frequency exponent is less than one and decreases with temperature, which is characteristic of hopping conduction. The temperature dependence of DC conductivity was examined to identify thermally activated behavior with realistic activation energy. Impedance spectroscopy was employed to distinguish bulk hopping contributions from grain boundary effects. Additionally, structural and spectroscopic analyses were conducted to verify the presence of cation disorder or mixed valence states that facilitate polaron hopping. Collectively, these methods provide a comprehensive assessment of whether polaron hopping constitutes the dominant conduction mechanism.

3.4. DC conductivity and activation energy

Fig. 8 presents Arrhenius plots of dc conductivity for Se-substituted Sr-NHFs with varying substitution ratios ($x \leq 0.10$). The approximately linear regions at lower $1/k_B T$ (higher temperatures) indicate thermally activated conduction consistent with an Arrhenius-type behavior. The observed slopes reflect activation energies for conduction, with slight variations among different Se contents. In general, samples exhibit decreasing activation energies with increasing temperature, suggesting enhanced carrier mobility at elevated temperatures.

Additionally, the modest effect of substitution ratio on the activation energy implies that Se incorporation modifies defect chemistry and local electronic structure without fundamentally altering the thermally activated hopping mechanism. This behavior is typical of ferrite systems where substituent ions subtly influence charge transport by modifying cation distributions or defect states.

Studies have shown that both E_a and DC conductivity fluctuate with different Se-ion substitution ratios in Sr-NHFs. A notable deviation occurs at the substitution ratio of $x = 0.06$, where the changes are more pronounced compared to other ratios. While the curvature variations in DC conductivity exhibit similar trends across different substitution ratios, the fluctuations are distinct, reflecting the impact of substitution levels.

At higher temperatures ($T > 70^\circ\text{C}$), E_a increases, whereas at lower temperatures ($T < 70^\circ\text{C}$), it decreases and, in some cases, becomes negative, depending on the Se-ion substitution ratio. Importantly, across both temperature ranges, the highest E_a is consistently observed at $x = 0.06$. This suggests that the substitution ratio plays a critical role in determining the E_a and conductivity behavior of Sr-NHFs.

Understanding the DC conductivity and E_a of Se→Sr NHFs requires an analysis of how Se incorporation influences electronic transport mechanisms, and the energy required for charge carriers to move through the ferrite material. In hexaferrites, DC conductivity primarily arises from electron hopping between Fe^{2+} and Fe^{3+} ions at Oh sites. This charge transport mechanism is affected by temperature, the nature of the substituent, and the microstructure. The introduction of Se ions modifies charge distribution, local electric fields, and structural symmetry, thereby influencing the ease of electron hopping between sites. Therefore, when Se ion is substituted into Sr-NHFs, it may occupy either Td or Oh sites, leading to changes in lattice parameters, bond lengths, and bond angles. These structural modifications alter the electronic band structure, affecting the density of states and the mobility of charge carriers. As a chalcogen, Se can introduce new energy levels within the bandgap or influence existing levels associated with Fe ions, thereby enhancing or suppressing DC conductivity depending on its effect on the electron hopping mechanism.

E_a represents the minimum energy required for charge carriers to

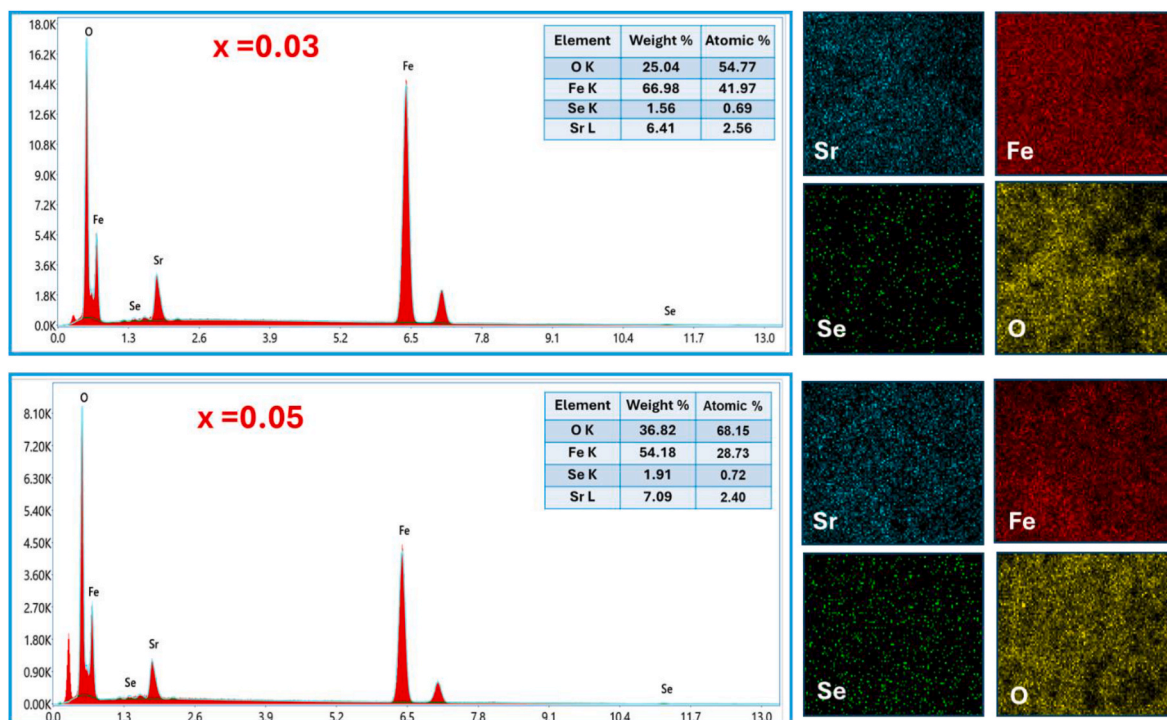


Fig. 3. EDX spectra and elemental mapping images of Se→Sr NHFs ($x = 0.03$ and 0.05).

hop from one site to another. In terms of DC conductivity, a lower E_a indicates easier charge carrier movement, generally leading to higher conductivity. Se substitution modifies E_a by altering potential barriers for electron hopping. Experimentally, E_a is determined from the temperature dependence of DC conductivity using the Arrhenius equation (Equation (3)) [33,34]:

$$\sigma(T) = \sigma_0 \exp\left(-\frac{E_a}{k_B T}\right) \quad (3)$$

where σ is the DC conductivity at temperature T , σ_0 is the pre-exponential factor, k_B is the Boltzmann constant, and T is the absolute temperature in K. Existing literature on ferrites suggests that DC conductivity typically increases with temperature, exhibiting Arrhenius-type behavior [24,35,36]. For Se-substituted Sr-NHFs, our findings indicate a reduction in E_a compared to unsubstituted Sr-NHFs, suggesting that Se facilitates easier electron hopping between Fe ions [37]. The E_a , extracted from the Arrhenius relation, fluctuates with increasing Se content, further indicating that Se can enhance charge carrier hopping. Prior research on other substituents such as Zn, NbY, Gd, Mo, and Ti in Sr-NHFs [24,27,32,38] has shown that these elements can either enhance or suppress DC conductivity and E_a , depending on their electronic configuration and the resulting structural changes. Se's unique electronic structure introduces distinct modifications in these properties, necessitating comparisons with similar studies for a more comprehensive understanding.

Microstructural factors, particularly grain size and grain boundaries,

significantly influence DC conductivity. Smaller grains tend to increase the number of grain boundaries, which act as barriers to charge carrier movement, potentially raising E_a . Higher grain boundary density may reduce DC conductivity by limiting the effective cross-sectional area available for charge carriers and increasing scattering events.

Previous studies on Se substitution in other ferrites suggest that Se can introduce localized states within the bandgap, affecting both DC conductivity and E_a . The specific impact on Sr-NHFs depends on how Se modifies Fe–O–Fe bonds and alters the overall electronic structure. Temperature-dependent DC conductivity measurements provide valuable insights into the structural and electronic changes induced by Se substitution [37,39].

Comparing these results with other substituted Sr-NHFs (e.g., NbY, Ti, Zn) [24–27,32,37–39] can help elucidate the unique effects of Se on DC conductivity and E_a . Identifying the optimal Se substitution level that enhances DC conductivity while minimizing E_a is crucial for practical applications in electronic devices. Improved DC conductivity and reduced E_a could make Se→Sr NHFs suitable for applications such as magnetic sensors, inductors, and electronic components where high conductivity is advantageous. This study offers a comprehensive analysis of how Se substitution influences the DC conductivity and E_a of Sr-NHFs, supported by relevant literature on ferrites and similar materials [37,39].

3.5. Dielectric constant

The dielectric constant of Se→Sr NHFs exhibits strong dependencies

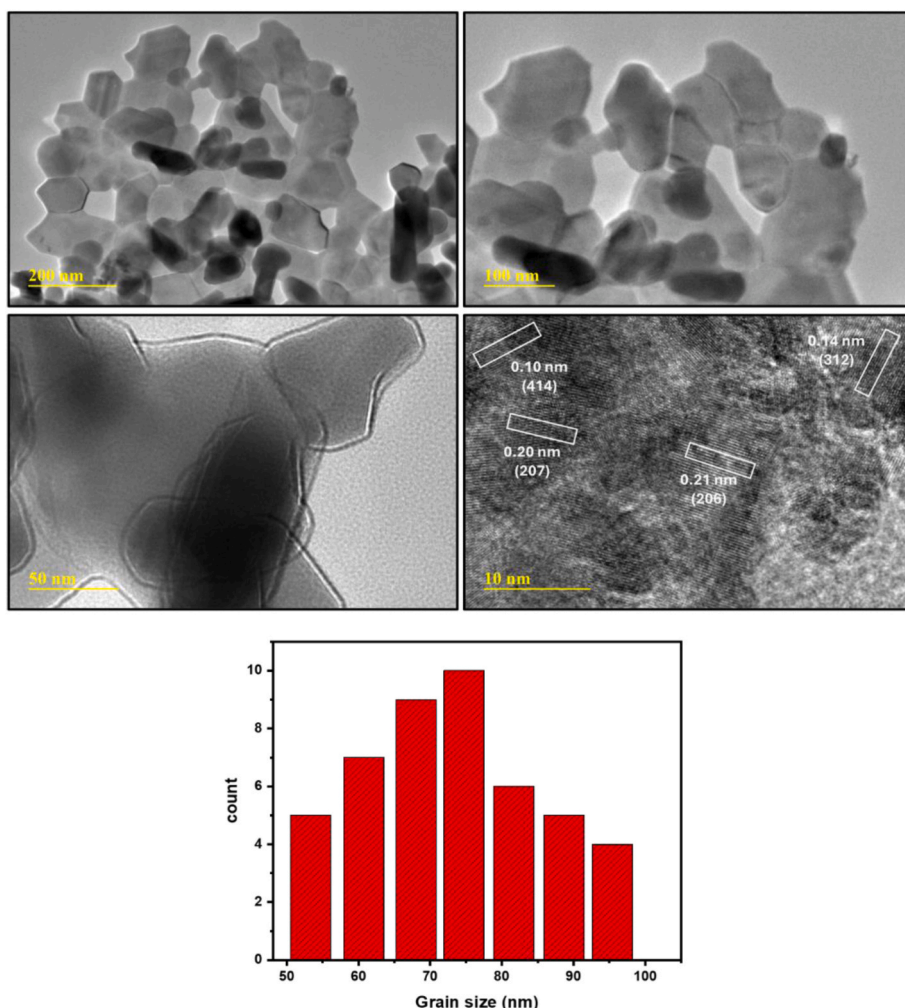


Fig. 4. TEM, HR-TEM images, and grain size distribution of Se→Sr ($x = 0.05$) NHFs.

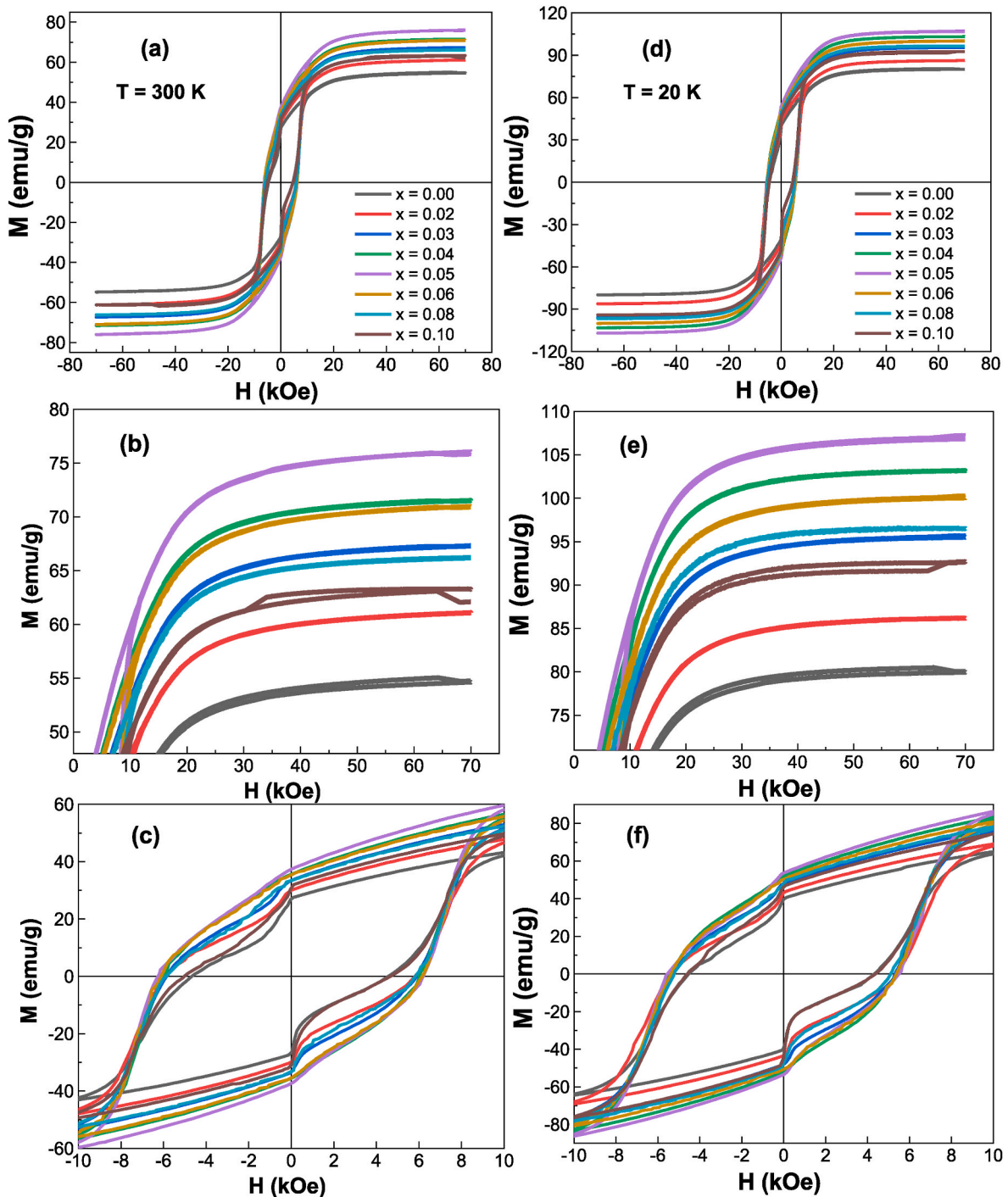


Fig. 5. $M - H$ curves for Se→Sr NHFs measured at (a–c) $T = 300$ K and (d–f) $T = 20$ K.

on frequency, temperature, and substitution ratio, playing a critical role in determining their capacitive performance and polarization behavior. Across all substitution levels, the dielectric constant generally decreases with increasing frequency as seen in Fig. 9, a trend commonly explained by the Maxwell-Wagner polarization model. At lower frequencies, multiple polarization mechanisms—including space charge, dipolar, ionic, and electronic polarization—contribute significantly to the dielectric response [22,40]. However, at higher frequencies, only faster polarization mechanisms, such as electronic polarization, remain active, leading to a reduced dielectric constant. Notably, a sudden drop in dielectric constant is observed at $x = 0.01$ at low temperatures and high frequencies, while other substitution ratios do not exhibit this abrupt

variation. Despite variations in substitution levels, the dielectric constant tends to remain relatively stable except for this anomaly at $x = 0.01$. Additionally, the maximum dielectric constant is consistently observed at high temperatures and low frequencies, where thermal activation enhances charge carrier mobility and polarization effects.

Se ion substitution into Sr-NHFs modifies the material's microstructure and local electric fields, influencing charge carrier dynamics and polarization mechanisms. Se ions, existing in multiple oxidation states (Se^{2-} , Se^{4+} , Se^{6+}), can replace oxygen or occupy interstitial sites within the lattice, modifying charge distribution, lattice parameters, and defect density. These modifications affect polarization behaviors, including interfacial polarization at grain boundaries, which is

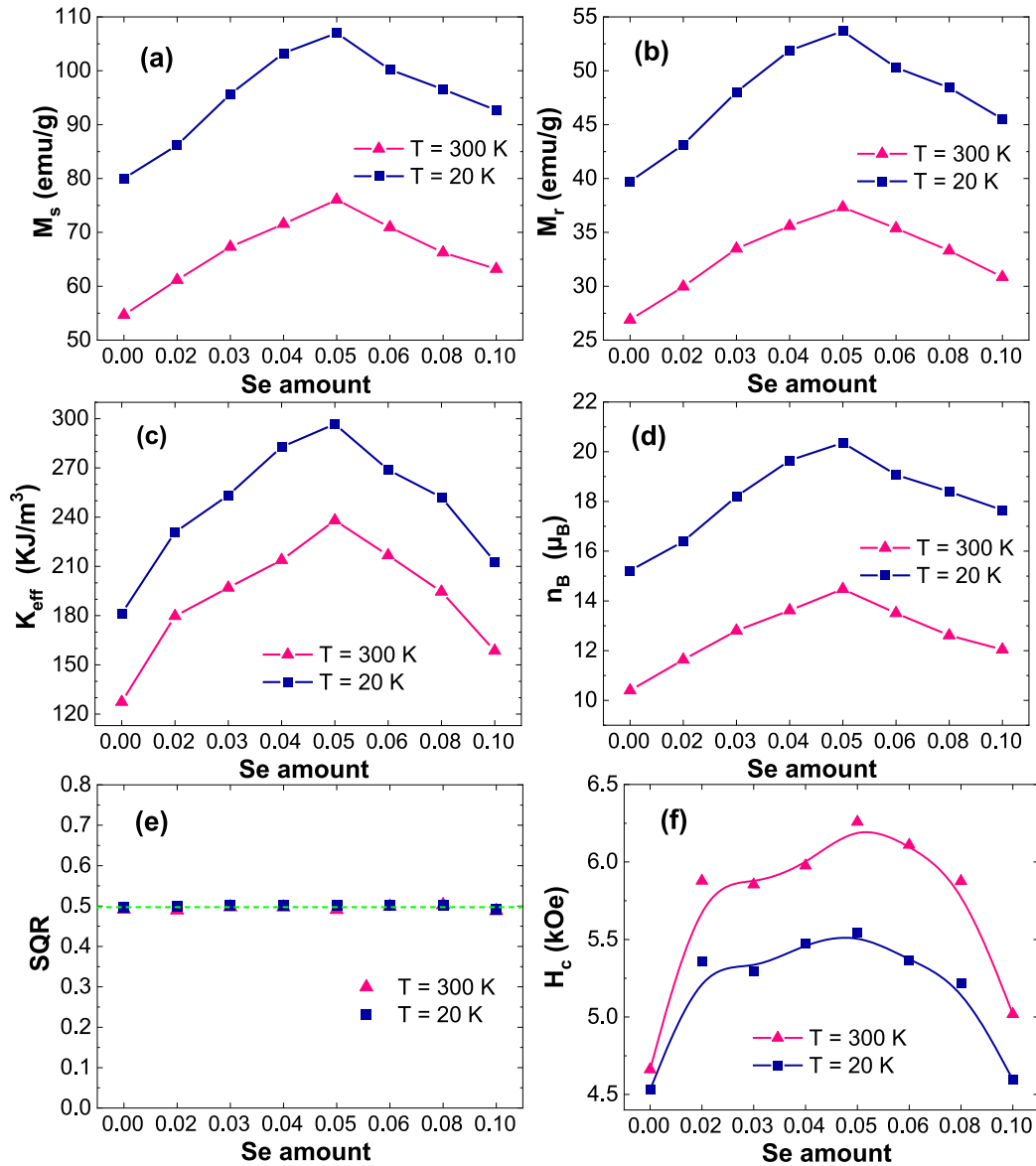


Fig. 6. Dependence of (a) M_s , (b) M_r , (c) K_{eff} , (d) n_B , (e) SQR, (f) H_c with Se amount.

particularly significant in heterogeneous ferrite materials. Furthermore, Se incorporation may shift relaxation frequencies, thereby impacting the dielectric response across different frequency ranges. The temperature dependence is also influenced by Se substitution as seen in the relevant graphs, as increased thermal agitation facilitates dipole alignment with the applied electric field. If Se substitution induces phase transitions in Sr-NHFs, these transitions can lead to sharp changes in the dielectric constant due to differing polarization capabilities in each phase.

Comparative studies with other substituted ferrites (e.g., NbY, Zn, Gd, Mo, and Ti) suggest that substituent ions impact the dielectric constant through changes in microstructure, grain boundary density, and ionic radius effects [16,17,19,32,38]. The specific electronic configuration of Se may introduce localized states within the bandgap, modifying space charge polarization and influencing overall dielectric behavior. Additionally, smaller grains with higher boundary density often exhibit enhanced dielectric constants due to increased interfacial polarization. Measuring dielectric constant and dielectric loss across various frequencies provides insights into the dominant polarization mechanisms and relaxation processes. By optimizing Se substitution levels, Sr-NHFs can achieve enhanced dielectric properties, making

them suitable for high-frequency applications, energy storage systems, and microwave components.

3.6. Dielectric loss

The dielectric loss of Se→Sr NHFs exhibits strong dependence on frequency, temperature, and substitution ratio, reflecting the material's energy dissipation characteristics in an alternating electric field as seen in [Supplementary Fig. S1](#). Notably, at $x = 0.01$, a sudden peak in dielectric loss occurs at low temperatures and medium frequencies, while no similar trend is observed for other substitution levels. Despite variations in substitution ratios, dielectric loss remains relatively constant, except for this anomaly. At low frequencies, different substitution levels exhibit varied trends across the entire temperature range, whereas at high frequencies, dielectric loss follows a more uniform pattern. The maximum dielectric loss for all substitution levels is typically observed at high temperatures and low frequencies, similar to the dielectric constant's behavior. In general, dielectric loss decreases and then increases with frequency, though the specific tendencies depend on the substitution ratio.

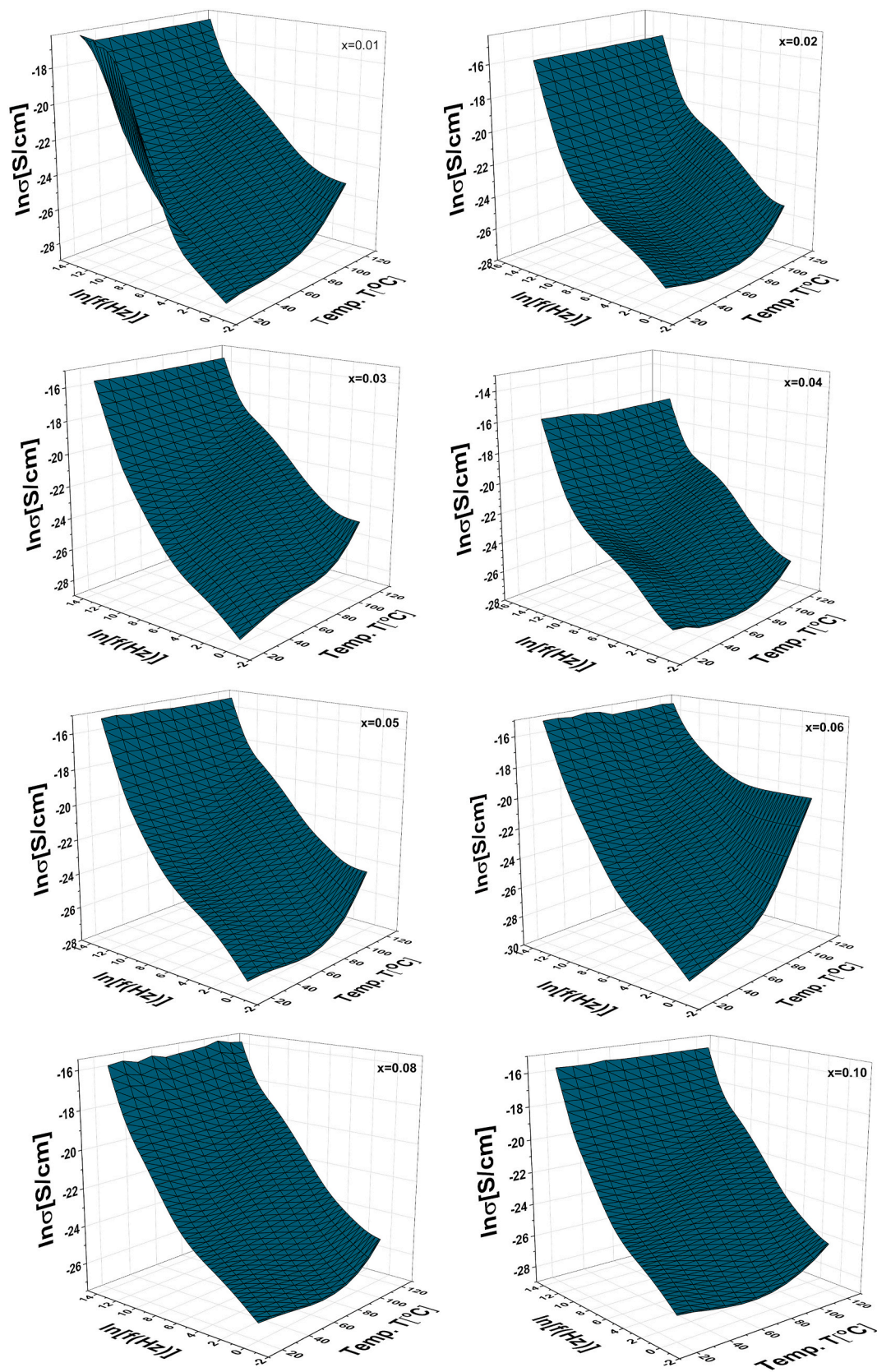


Fig. 7. 3D plot of the f - and T -dependent conductivity of Se→Sr NHFs (See Ref. [3] for $x = 0.00$).

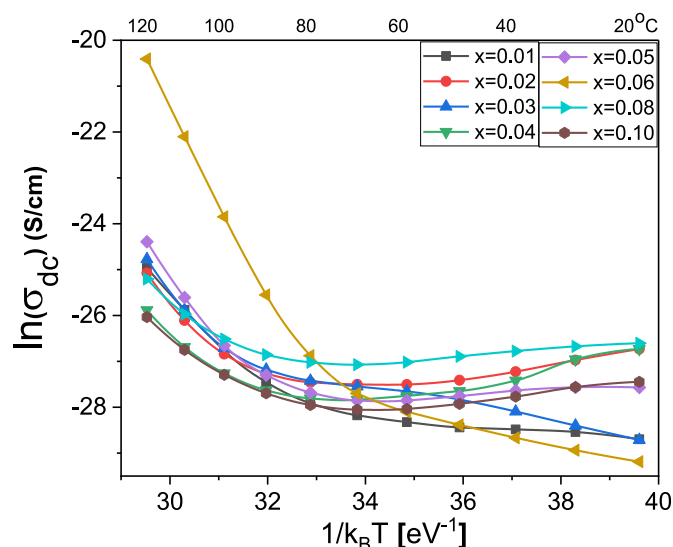


Fig. 8. Arrhenius plot of dc conductivity of Se→Sr NHFs ($x \leq 0.10$) (See Ref. [3] for $x = 0.00$).

Dielectric loss in ferrites arises from multiple mechanisms, including ionic and dipolar relaxation (*delays in dipole alignment and ion displacement*), conductivity losses (*charge carrier movement*), and interfacial or space charge polarization (*charge trapping at grain boundaries*). Typically, dielectric loss increases with temperature due to enhanced dipole relaxation and charge carrier mobility, but decreases at higher frequencies as slower polarization mechanisms become less effective. Se ion substitution significantly influences these dielectric loss mechanisms by modifying the crystal structure and electronic environment of Sr-NHFs. Se ions, capable of existing in multiple oxidation states (Se^{2-} , Se^{4+} , Se^{6+}), may replace oxygen or occupy interstitial sites, introducing localized states and changing defect states within the bandgap. These changes affect charge carrier dynamics and polarization processes, thereby influencing dielectric loss. Specifically, Se substitution impacts: (i) Se may stabilize or destabilize dipoles, leading to either enhanced or reduced dielectric loss, (ii) Changes in grain boundary density due to Se substitution can influence space charge polarization and dielectric loss and (iii) Finally, the effects of Se ion on the electronic structure can modify DC conductivity, directly impacting dielectric loss through energy dissipation during charge carrier movement.

Studies on NbY-, Mo-, Ti-, and Al-substituted ferrites indicate that substituent ions can either increase or decrease dielectric loss, depending on their effects on polarization and conduction processes [24,27,32,41]. Therefore, Se substitution introduces unique outcomes due to its electronic configuration and bonding characteristics. As frequency increases, dielectric loss generally declines since slower polarization mechanisms (*such as ionic and space charge polarization*) lose effectiveness. However, Se substitution may shift relaxation frequencies, modifying the dielectric loss profile across different frequency ranges. Additionally, rising temperatures typically lead to higher dielectric loss due to increased thermal activation of dipoles and charge carriers, and Se substitution could alter the E_a 's of these processes, affecting the temperature dependence of dielectric loss.

Microstructural properties, particularly grain size and boundary density, play a crucial role in dielectric loss. Se substitution may influence grain size distribution, increasing grain boundary areas and consequently enhancing space charge polarization. Increased porosity, which introduces charge trapping sites and scattering effects, can further contribute to higher dielectric loss. Comparative analysis with NbY-, Mo-, Co-, and Ti-substituted ferrites provides insight into the specific effects of Se on dielectric loss, highlighting differences in ionic radius, electronic structure, and bonding characteristics [20–27,32,41]. The

ability to optimize Se substitution levels to balance low dielectric loss with a high dielectric constant is critical for applications such as capacitors, inductors, and high-frequency devices. Reducing dielectric loss while maintaining desirable dielectric properties enhances the efficiency of Sr-NHFs in energy storage and microwave applications. By integrating findings from relevant literature, this analysis presents a comprehensive understanding of the impact of Se substitution on dielectric loss in Sr-NHFs, considering structural, electronic, and microstructural influences.

3.7. Dissipation factor

The dissipation factor, also known as the loss tangent ($\tan \delta$), is a crucial parameter in evaluating the dielectric behavior of Se-substituted Sr-NHFs. It represents the ratio of energy dissipated as heat to the energy stored when subjected to an alternating electric field, quantifying the material's inefficiency in storing electrical energy. A lower dissipation factor indicates improved performance for applications requiring minimal energy loss, such as capacitors and high-frequency electronic devices.

The dissipation factor in Sr-NHFs exhibits strong dependence on frequency, temperature, and substitution ratio as demonstrated in Fig. 10. Notably, at $x = 0.01$, a broad peak in the dissipation factor appears at low temperatures and medium frequencies, while no such trend is observed for other substitution levels. At low frequencies, different substitution levels show varied trends across the entire temperature range, whereas at high frequencies, the dissipation factor follows a more uniform pattern. The maximum dissipation factor for all substitution ratios typically occurs at high temperatures and low frequencies, similar to the behavior of the dielectric loss. The most significant variations in the factor appear at low frequencies and high temperatures, particularly for $x = 0.06$, with similar trends recorded for $x = 0.02, 0.04, 0.05, 0.08$, and 0.10 . In general, the dissipation factor decreases and then increases with frequency, though its specific behavior varies depending on the substitution ratio.

The dissipation factor in ferrites arises from multiple loss mechanisms, including: (i) Energy loss due to a lag in polarization response to the applied electric field (*polarization losses*), (ii) Movement of free charge carriers (e.g., electrons and ions), leading to resistive heating (*conductivity losses*), and (iii) Charge accumulation at grain boundaries, a significant factor in heterogeneous materials like ferrites (*interfacial polarization*). As frequency increases, the dissipation factor generally decreases because slower polarization mechanisms, such as interfacial polarization, become less effective. At lower frequencies, these mechanisms dominate, leading to higher dielectric loss and an increased dissipation factor. Se substitution may shift the relaxation frequencies of these polarization processes, altering the frequency dependence of the dissipation factor. Similarly, temperature has a direct impact on the dissipation factor, which typically increases as temperature rises due to enhanced thermal motion that amplifies dielectric loss. Se substitution may modify this temperature dependence by altering the E_a required for charge carrier movement and polarization processes.

Se substitution into Sr-NHFs induces structural modifications, including changes in lattice parameters, symmetry, and ion distribution, which can significantly influence polarization mechanisms and, consequently, the dissipation factor. Depending on its oxidation state and site occupancy, Se may introduce localized electronic states within the bandgap, which interact with the applied electric field and affect both dielectric loss and the dissipation factor. Key ways in which Se substitution impacts the dissipation factor include: (i) Se substitution can alter the mobility and concentration of charge carriers (electrons and holes), affecting the material's conductivity. Increased conductivity often leads to a higher dissipation factor due to greater energy dissipation as heat (*Charge Carrier Dynamics*). (ii) Se substitution influences grain size and boundary density. A higher grain boundary density enhances interfacial polarization, raising the dissipation factor (*Grain Boundary Effects*). (iii)

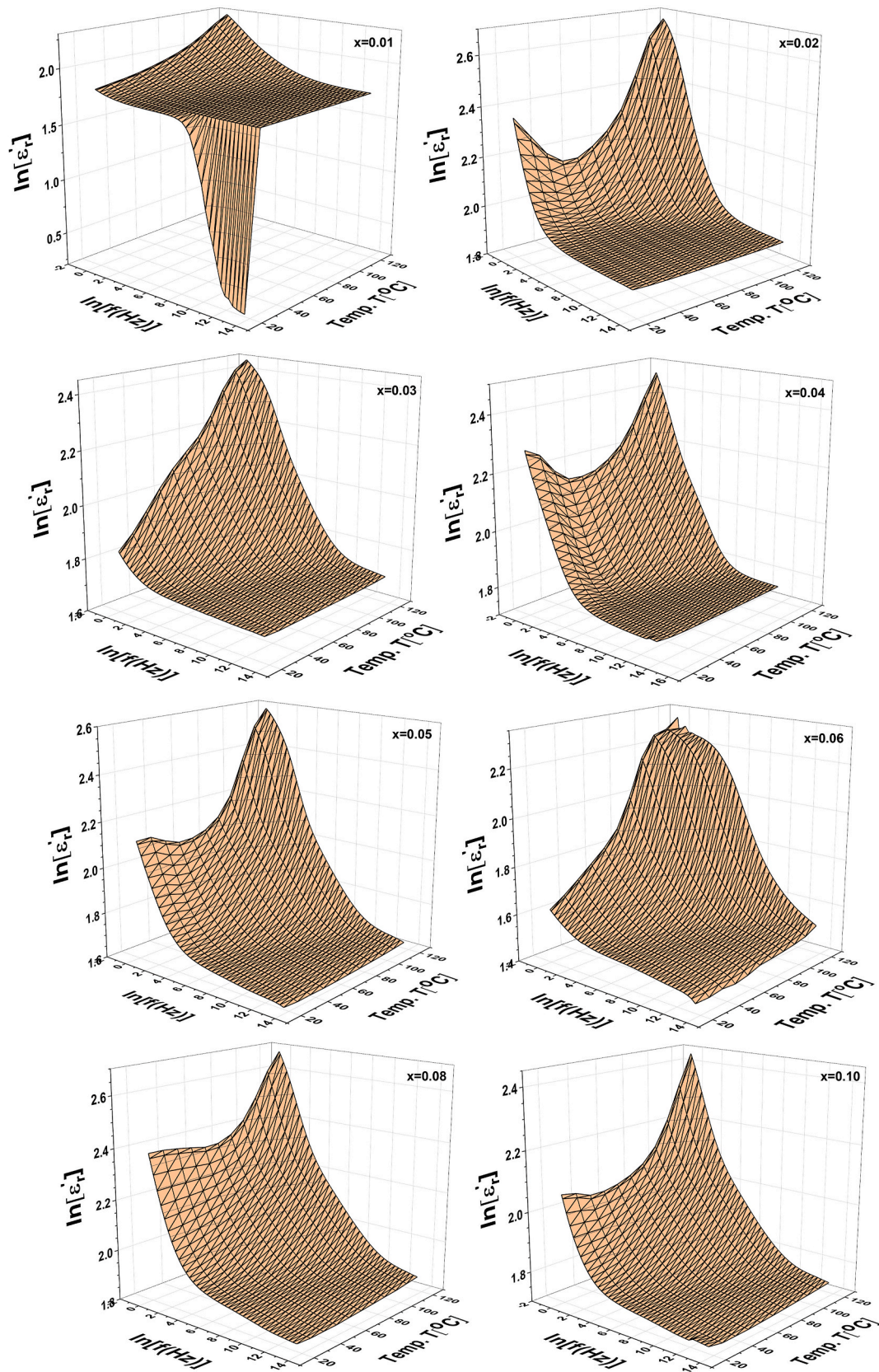


Fig. 9. 3D plot of the f - and T -dependent dielectric constant of Se→Sr ($x \leq 0.10$) NHFs (See Ref. [3] for $x = 0.00$).

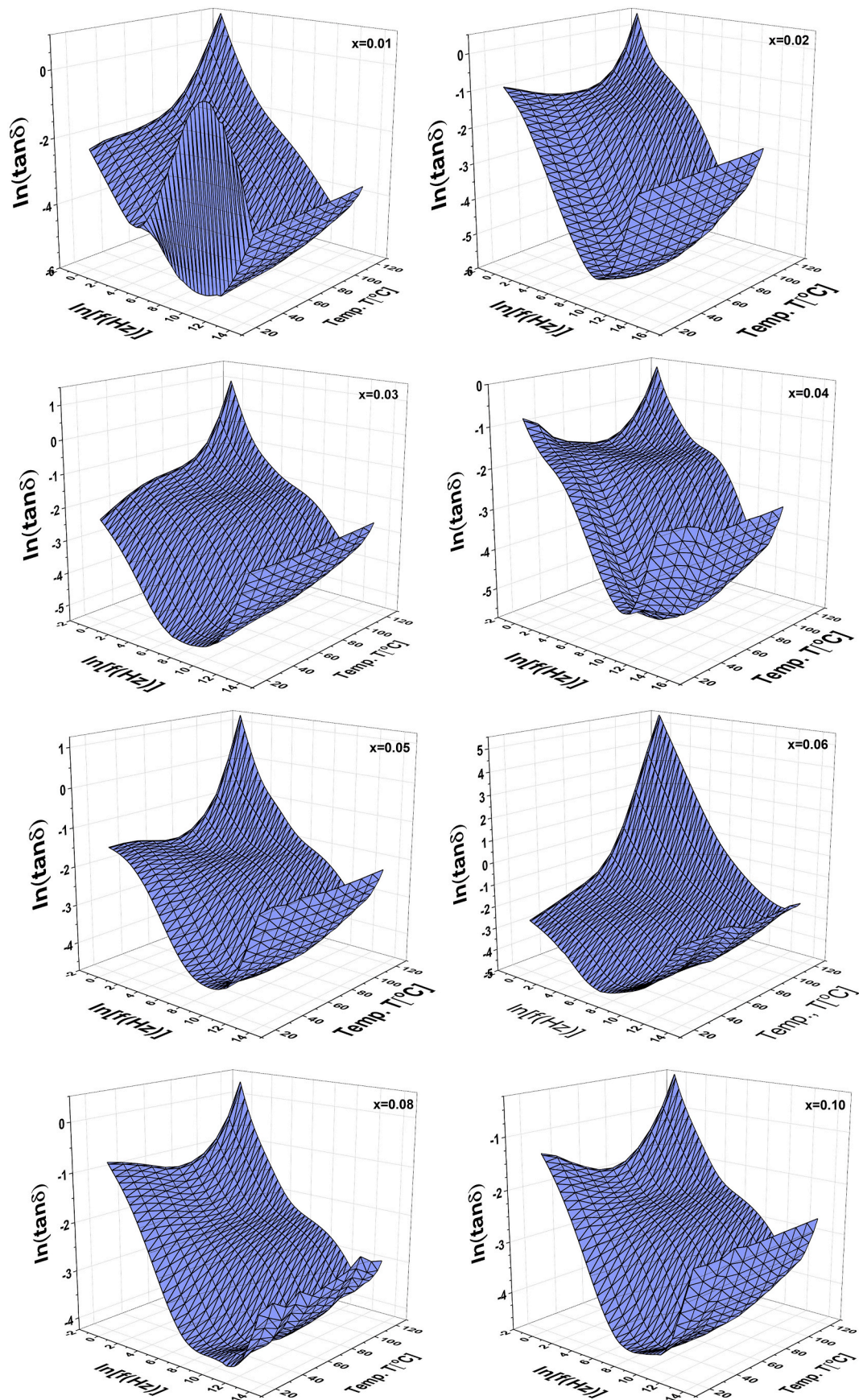


Fig. 10. 3D plot of the f - and T -dependent dissipation factor of Se→Sr ($x \leq 0.10$) NHFs (See Ref. [3] for $x = 0.00$).

Variations in grain size and porosity impact the dissipation factor. Increased porosity introduces additional charge trapping sites and scattering centers, elevating the dissipation factor (*Microstructural Changes*).

Studies on ferrites substituted with Mo, Tb–Tm, Nb–Y, Ti, V, and Ce reveal that the dissipation factor varies significantly based on the substituent, primarily due to changes in microstructure, grain boundary effects, and electronic structure [23–25,27,42,43]. Se's unique electronic configuration and its ability to introduce states within the bandgap may result in distinct effects compared to other substituents. Higher Se concentrations could increase localized defect states, leading to higher dielectric loss and an elevated dissipation factor. However, at optimized Se levels, the material's energy storage capacity may improve while minimizing losses, leading to a reduced dissipation factor.

3.8. Complex electrical modulus analysis

Analyzing the complex electrical modulus of Se→Sr NHFs provides valuable insights into the material's dielectric relaxation and conduction mechanisms. The complex modulus (M^*), composed of a real component (ReM) and an imaginary component (ImM), is defined as:

$$M^* = \text{Re}M + j\text{Im}M = \frac{1}{\epsilon^*} \quad (4)$$

where ϵ^* represents the complex permittivity (Equation (4)). Modulus analysis is particularly useful for distinguishing between grain and grain boundary contributions in polycrystalline materials such as ferrites, offering a more detailed understanding of relaxation processes. The real part of the electrical modulus (ReM) typically increases with frequency, especially at higher frequencies, where the polarization response becomes limited, as seen in [Supplementary Fig. S2](#). In Se→Sr NHFs, this behavior is influenced by structural changes and charge redistribution within the lattice due to Se incorporation. A higher ReM at high frequencies suggests restricted polarization mechanisms, likely due to modifications in grain boundary characteristics or the introduction of defect states. At $x = 0.01$, ReM exhibits a broad peak at low temperatures and medium frequencies, while no similar trend is observed for other substitution levels. ReM is highly dependent on substitution ratios, remaining almost constant except for a sudden increase at $x = 0.01$. At low frequencies, different substitution levels display distinct trends across the entire temperature range. However, at high frequencies, ReM follows a more uniform pattern across all samples. The most significant variations in ReM occur at high temperatures and low frequencies. The highest variations are observed at $x = 0.06$ under low-frequency and high-temperature conditions, with similar trends recorded for $x = 0.02, 0.04, 0.05, 0.08$, and 0.10 . In general, ReM increases with frequency for all substitution levels, although the rate of increase varies with composition.

The imaginary part of the electrical modulus (ImM) represents energy loss associated with relaxation processes as depicted in [Supplementary Fig. S3](#). The presence of peaks in ImM indicates relaxation events, with the peak position corresponding to the characteristic relaxation time given by the equation (Equation (5)):

$$\tau = \omega^{-1} \quad (5)$$

where ω is the angular frequency at the peak. The presence of multiple peaks in the ImM spectrum suggests the coexistence of multiple relaxation mechanisms as in the case of $x = 0.06$. These could arise from variations in the local environment due to Se substitution, leading to a distribution of relaxation times influenced by grain and grain boundary effects. Se substitution can lower the energy barrier for ion hopping or modify the relaxation frequency, causing shifts in ImM peaks toward higher frequencies. This indicates faster relaxation dynamics, potentially due to alterations in lattice parameters, defect states, and charge carrier behavior. At $x = 0.01$, ImM displays a broad peak at low

temperatures and medium frequencies, with no comparable trend observed for other substitution levels. ImM is highly dependent on substitution ratios, except for a sudden increase at $x = 0.01$. At low frequencies, different substitution levels exhibit distinct behaviors across the temperature range, while at high frequencies, ImM follows a more uniform trend. In most cases, ImM displays two or more peaks at low and high temperatures under low-frequency conditions. However, at $x = 0.06$, four peaks emerge at high temperatures and low frequencies. For $x = 0.02, 0.03, 0.05$, and 0.08 , ImM exhibits two peaks under these conditions. The highest variations in ImM occur at high temperatures and low frequencies, with the most pronounced effects observed at $x = 0.06$, followed by $x = 0.02, 0.04, 0.05, 0.08$, and 0.10 . Similar to ReM, ImM increases with frequency, though the specific trend varies depending on the substitution ratio.

The nature of relaxation in Se→Sr NHFs can be classified as either Debye or non-Debye based on the ImM spectra: (i) *Debye relaxation* characterized by a single, symmetric peak in ImM, indicating a uniform relaxation time distribution. (ii) *Non-Debye relaxation* exhibited by broad or asymmetric ImM peaks, suggesting multiple relaxation mechanisms or a distribution of relaxation times. Se substitution is likely to induce non-Debye behavior by modifying defect states and grain boundary effects, leading to a broader relaxation time distribution [33,44].

The complex modulus spectrum provides insights into the contributions of grain and grain boundary effects: firstly, high-frequency region is typically associated with grain (bulk) effects. So, an increase in ReM and ImM at high frequencies suggests that Se substitution has modified bulk properties, likely through changes in charge carrier mobility or lattice strain. Secondly, low-frequency region is dominated by grain boundary effects. If Se substitution increases grain boundary density or introduces new defect states, additional relaxation peaks may appear in the low-frequency range, indicating enhanced energy dissipation due to grain boundary polarization [45]. Understanding the complex modulus behavior of Se-substituted Sr-NHFs is crucial for optimizing their performance in electronic applications. By analyzing ReM and ImM, the optimal Se substitution level can be determined to balance conductivity and dielectric relaxation. The complex electrical modulus analysis of Se-substituted Sr-NHFs reveals that Se incorporation significantly impacts dielectric relaxation dynamics, polarization mechanisms, and conduction processes. By tailoring Se substitution levels, Sr-NHFs can be optimized for high-frequency and energy-sensitive applications. The insights gained from this analysis not only clarify the role of Se in modifying ferrite behavior but also offer guidelines for developing advanced functional materials for electronic and microwave technologies [46].

3.9. Cole-Cole plot analysis of complex electrical modulus

The Cole-Cole plot is a widely used tool in dielectric spectroscopy to analyze the relaxation behavior of materials by plotting the imaginary part of the modulus ($-\text{Im}M$) versus the real part (ReM). This approach helps in understanding the distribution of relaxation times and distinguishing between Debye and non-Debye relaxation processes in Se→Sr NHFs. So, the Cole-Cole plot typically forms semi-circular arcs, where each arc corresponds to a different relaxation process. The shape and symmetry of these arcs provide information about the nature of dielectric relaxation in the system: Firstly, a single symmetric semicircle indicates Debye relaxation, where there is a single, well-defined relaxation time. Secondly, a distorted or depressed semicircle suggests non-Debye relaxation, where multiple relaxation processes contribute, leading to a broad distribution of relaxation times. Thirdly, multiple semicircles reveal the presence of grain and grain boundary effects, with separate arcs corresponding to bulk/grain and interfacial relaxation mechanisms as demonstrated in [Fig. 11](#). Therefore, the Cole-Cole plot for Se-substituted Sr-NHFs does not exhibit a perfect semicircle, indicating non-Debye relaxation behavior. The deviation from an ideal semicircle suggests the presence of multiple relaxation mechanisms,

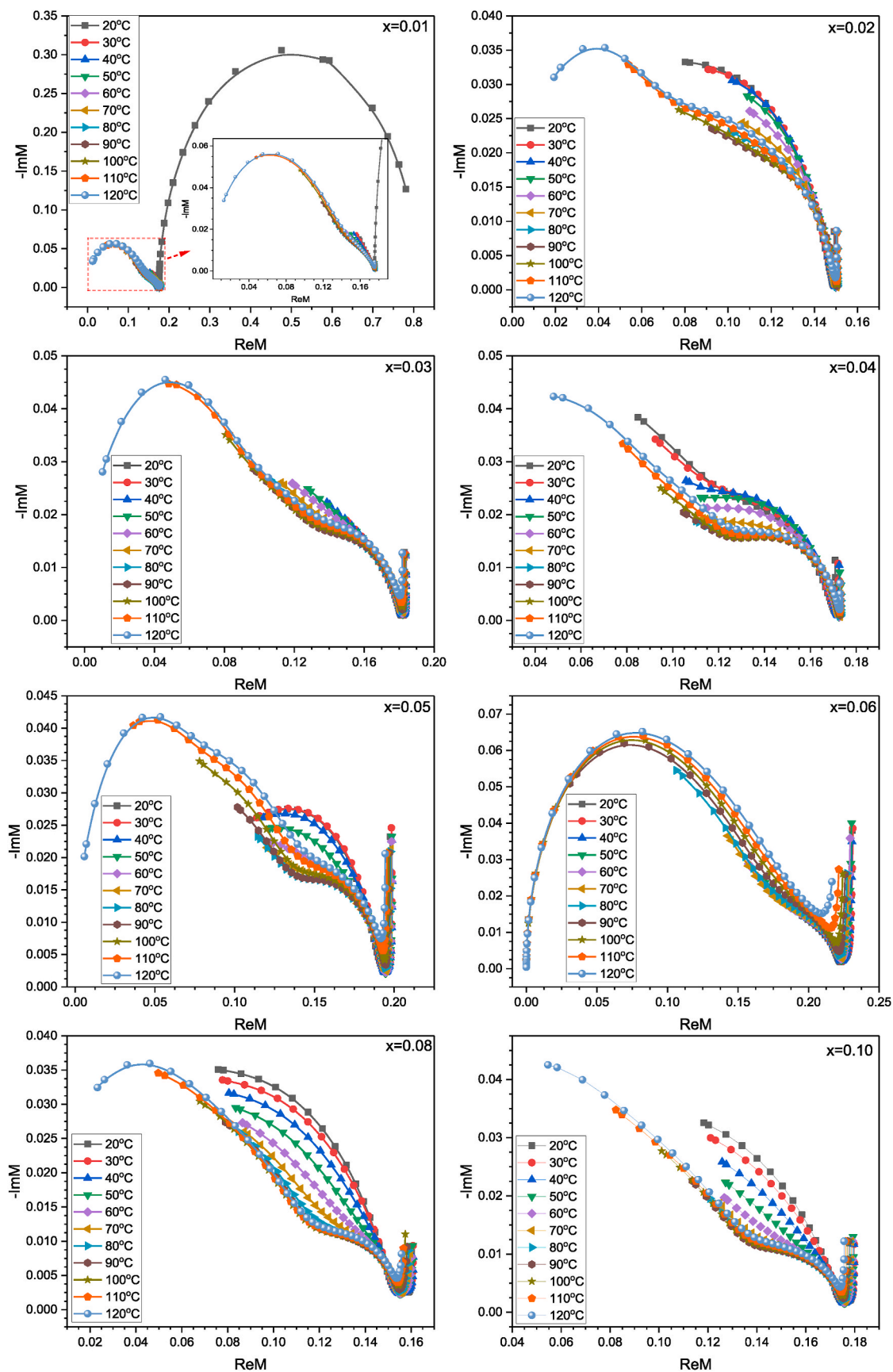


Fig. 11. Cole-cole plot of complex electrical modulus of Se→Sr NHFs.

which arise due to the introduction of Se ions modifying the local lattice environment. Se substitution leads to changes in defect states, charge carrier dynamics, and grain boundary characteristics, resulting in a broad distribution of relaxation times rather than a single well-defined relaxation time.

The radius of the semicircle varies with substitution levels, indicating that different Se concentrations influence the dielectric relaxation strength. At low substitution levels ($x = 0.01$ to $x = 0.04$), the Cole-Cole plot shows a larger arc, suggesting dominant bulk relaxation effects. At higher substitution levels ($x = 0.06$ to $x = 0.10$), the arcs become more depressed and elongated, indicating increasing grain boundary contributions and defect-induced relaxation. The shift of the arcs toward higher ReM values at increased Se concentrations implies enhanced resistive behavior and a reduction in charge carrier mobility. At low frequencies, the plot shows a higher curvature and larger arc due to the dominance of space charge polarization and grain boundary effects. The presence of multiple relaxation processes is more evident at lower frequencies, where charge carriers have more time to respond to the applied field. At high frequencies, the arc shrinks, indicating that the bulk effects dominate as grain boundary contributions become less significant. The material's response becomes more resistive, leading to a shift in the relaxation peak position toward higher frequencies. At high temperatures, the semicircles become more distorted, confirming that thermally activated charge carrier hopping plays a crucial role in relaxation dynamics. A shift in relaxation frequency to lower values is observed, consistent with thermally activated conduction mechanisms. The broad and depressed semicircles as seen in the relevant graphs in Fig. 11 suggest that Se substitution induces a distribution of relaxation times, which may be beneficial for energy storage and high-frequency applications. By adjusting Se concentration, the balance between bulk/grain and grain boundary effects can be controlled, optimizing Sr-NHFs for specific applications such as microwave absorption, capacitors, and sensors [24]. Since grain boundaries significantly influence relaxation behavior, optimizing processing conditions (e.g., sintering temperature, grain size control) can further refine the dielectric properties of Se-substituted Sr-NHFs.

The Cole-Cole analysis of ImM-ReM for Se→Sr NHFs confirms that Se ion incorporation leads to non-Debye relaxation behavior, characterized by broadened and asymmetric semicircles in the modulus plot. The impact of Se substitution on relaxation dynamics is evident through variations in semicircle shape, relaxation frequency shifts, and increasing grain boundary contributions at higher substitution levels. These findings provide key insights into optimizing Sr-NHFs for high-frequency and energy-sensitive applications, where tailored dielectric properties are essential.

3.10. Cole-Cole plot analysis of complex impedance

The Nyquist plot, also known as the Cole-Cole plot, is an essential tool for analyzing the electrical properties of materials, particularly in understanding conduction mechanisms and relaxation processes. By plotting the imaginary part of impedance (ImZ) against the real part of impedance (ReZ), valuable insights can be obtained regarding charge transport, grain and grain boundary effects, as well as the overall resistive and capacitive behaviors of Se→Sr NHFs.

In electrical impedance spectroscopy, the Nyquist plot provides significant information about conduction mechanisms, relaxation processes, and resistive-capacitive behavior in Sr-NHFs, as shown in Fig. 12. In this study, we focus on Se→Sr NHFs, specifically examining firstly the temperature-dependent impedance from RT to 120 °C for $x = 0.02$ as depicted in Fig. 13(a) and secondly, the composition-dependent impedance for Se substitution ratios ranging from $x = 0.01$ to $x = 0.10$ at 20 °C as portrayed in Fig. 13(b). A typical Nyquist plot represents ImZ versus ReZ, where semicircular arcs correspond to relaxation processes within the material. The diameter of each semicircle relates to resistance contributions, including grain resistance (R_g) and grain

boundary resistance (R_{gb}). Additionally, a linear tail at low frequencies may indicate electrode polarization or ion diffusion effects.

Regarding temperature dependence for $x = 0.02$, as the temperature increases, the semicircle shrinks, indicating a decrease in total resistance (R_{tot}) due to thermally activated charge carriers. The plot shifts toward lower ReZ values, confirming an increase in electrical conductivity ($\sigma \propto 1/R$). Moreover, the relaxation frequency increases with temperature, meaning that charge carriers can respond more rapidly to an alternating field. Deviations from a perfect semicircle suggest non-Debye relaxation, which implies a distribution of relaxation times due to structural disorder introduced by Se substitution.

For composition dependence at 20 °C, the Nyquist plot exhibits different behaviors depending on the Se substitution level. At low Se substitution levels ($x = 0.01$ – 0.04), a single, well-defined semicircle is observed, suggesting that grain conduction dominates. However, for higher Se concentrations ($x = 0.06$ – 0.10), the plot features two overlapping semicircles, indicating distinct contributions from grains and grain boundaries. In the low-frequency region, increased impedance is evident for higher Se ion ratios, implying that Se substitution introduces defects, which in turn reduce mobility and increase grain boundary resistance. Furthermore, the increase in non-ideal behavior, characterized by semicircle distortion, suggests enhanced structural disorder and a broader range of relaxation times.

The general characteristics of a Nyquist plot in complex impedance analysis can be summarized as follows: initially, a single, well-defined semicircle signifies a single relaxation process, typically associated with grain conduction. Secondly, multiple semicircles indicate distinct conduction mechanisms, arising from both grains and grain boundaries. Thirdly, a depressed or incomplete semicircle suggests non-Debye relaxation, implying a broad distribution of relaxation times due to defects, charge trapping, or material heterogeneity. Lastly, a straight-line region at low frequencies is indicative of electrode polarization and ionic diffusion processes, which may result from space charge accumulation at grain boundaries.

The Cole-Cole plot of Se→Sr NHFs generally exhibits two overlapping semicircles, representing contributions from both grains and grain boundaries. The high-frequency semicircle is associated with intra-grain conductivity, reflecting the resistive-capacitive (RC) response of the grains. Meanwhile, the low-frequency semicircle arises due to grain boundary resistance, which is typically higher due to charge carrier trapping and interfacial polarization. The presence of distorted semicircles suggests non-Debye relaxation, implying a broad distribution of relaxation times as a result of defects introduced by Se substitution.

At low Se concentrations ($x = 0.01$ to 0.04), the high-frequency semicircle is more prominent, indicating that grain conduction dominates. This observation suggests that the material has relatively low grain boundary resistance, facilitating better charge transport. Conversely, at higher Se concentrations ($x = 0.06$ to 0.10), the low-frequency semicircle becomes more pronounced, signifying an increase in grain boundary resistance. This trend implies that Se ions introduce structural distortions, defects, or increased porosity, which hinder conduction by trapping charge carriers. At high frequencies, the impedance magnitude decreases, and the semicircle shifts toward lower ReZ values, indicating increased AC conductivity. The presence of a single semicircle at high frequencies suggests that grain effects dominate, whereas at low frequencies, a significant tail or additional semicircle emerges, highlighting the increasing role of grain boundary contributions and electrode effects. These characteristics are indicative of interfacial polarization and space charge effects, which are amplified by Se substitution.

To quantitatively analyze impedance behavior, the Nyquist plot is fitted using an equivalent circuit model. Typically, this model comprises (i) a grain resistance (R_g) and capacitance (C_g) in parallel to represent grain properties, (ii) a grain boundary resistance (R_{gb}) and capacitance (C_{gb}) in parallel to account for interfacial effects, and (iii) an additional

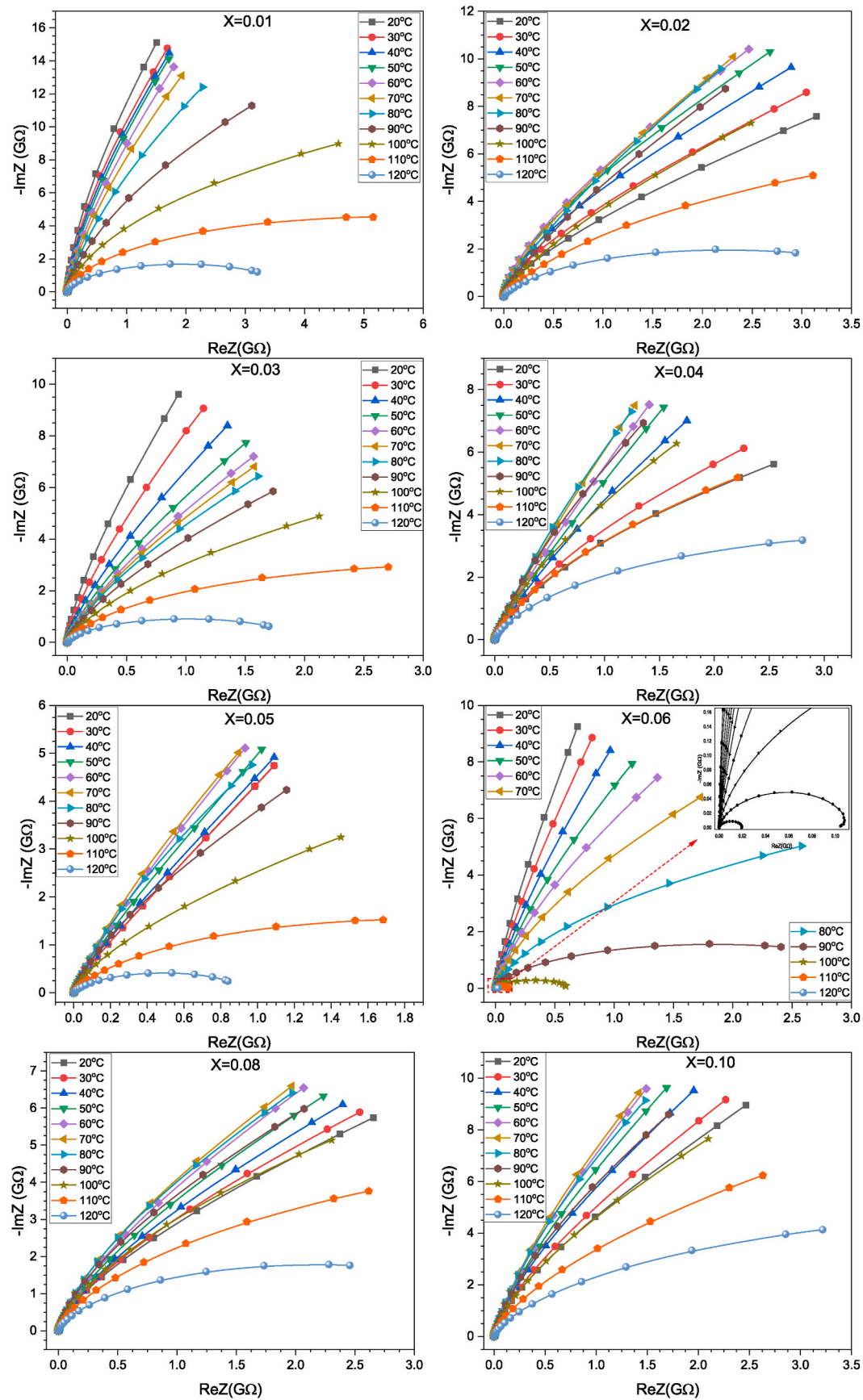


Fig. 12. Cole-Cole plot ($-\text{Im}Z$ vs. $\text{Re}Z$) of complex impedances for Se→Sr NHFs up to 3.0 MHz between 20 and 120 °C (See Ref. [3] for $x = 0.00$).

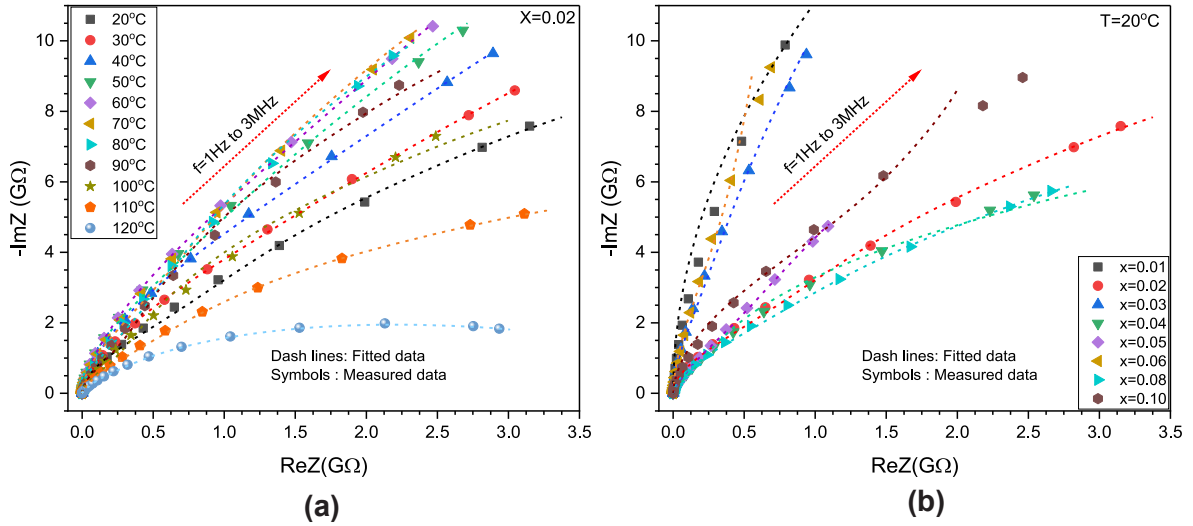


Fig. 13. Nyquist plot and fittings of the complex impedance for Se→Sr NHFs. The data covers (a) the temperature range from RT to 120 °C for $x=0.02$ and (b) the substitution ratio from 0.01 to 0.10.

constant phase element (CPE) to describe non-ideal dielectric behavior. The total impedance can be expressed as follows [47,48]:

$$Z = (R_g \parallel C_g) + (R_{gb} \parallel C_{gb}) + CPE \quad (6)$$

where R_g represents grain resistance, R_{gb} denotes grain boundary resistance, C_g and C_{gb} correspond to the capacitive responses of grains and grain boundaries, respectively, and CPE accounts for non-ideal capacitive behavior, indicating deviations from ideal Debye relaxation. As temperature increases, the semicircle radius decreases, signifying a reduction in total resistance ($R_g + R_{gb}$) due to thermally activated charge carrier conduction. The shift of the semicircle toward lower ReZ values confirms that conductivity follows an Arrhenius-type behavior, expressed as: $\sigma = \sigma_0 \exp(-E_a/k_B T)$. E_a decreases with increasing Se content, suggesting that Se substitution enhances charge carrier hopping. Additionally, the shift in relaxation frequency to higher values with temperature further supports the presence of thermally activated conduction.

For temperature dependence at $x = 0.02$, the total resistance ($R_{tot} = R_g + R_{gb}$) increases and then decreases with increasing temperature, confirming the existence of thermally-activated conduction mechanisms as listed in Tables 2 and 3. While the capacitance of grains (C_g) shows weak temperature dependence, the capacitance of grain boundaries (C_{gb}) slightly increases, suggesting enhanced polarization at grain boundaries. Regarding substitutional dependence at 20 °C, the grain resistance (R_g) decreases with increasing Se content up to $x = 0.04$. However, for higher Se concentrations, R_g increases, suggesting that excess Se introduces defects that trap carriers. Grain boundary resistance (R_{gb}) is highest for $x = 0.06-0.10$, indicating that Se substitution

increases grain boundary resistance due to structural disorder and charge trapping. Furthermore, the CPE behavior becomes more pronounced with increasing Se content, confirming non-ideal dielectric relaxation [49].

Overall, Nyquist plot fitting confirms the contributions of both grain and grain boundary effects in Se-substituted Sr-NHFs. Temperature dependence reveals thermally activated conduction, characterized by decreasing resistance and shifting relaxation frequencies. Se substitution influences impedance behavior, with compositions of $x = 0.02-0.04$ optimizing grain conduction, while $x = 0.06-0.10$ enhances grain boundary effects. Equivalent circuit modelling incorporating a CPE component suggests non-Debye relaxation, which is influenced by Se-induced structural disorder. The findings from Nyquist analysis demonstrate that Se substitution modifies conduction pathways and relaxation dynamics. The observed impedance trends, following Arrhenius-type behavior, can be adjusted by controlling Se concentration, making Se→Sr NHFs promising candidates for high-frequency, energy storage, and sensor applications [49,50].

3.11. Nyquist plot & its fittings for complex impedance

The Nyquist plot of Se→Sr NHFs is analyzed using the equivalent circuit model $R(CR)(QR)(CR)$ (Fig. S4). The fitting parameters of this model are evaluated for different temperatures ranging from 20 °C to 120 °C at a fixed substitution ratio of $x = 0.02$ (Table 2), as well as for varying Se ion substitution ratios (0.01–0.10) at a constant temperature of 20 °C (Table 3).

Key findings from the fitting analysis include: The Q square value is approximately 1.22. For $x = 0.02$, the characteristic frequency for grain

Table 2

Fitting parameters of the model $R(CR)(QR)(CR)$ of Nyquist plot of Se→Sr NHFs at temperatures between 20 and 120 °C for $x = 0.02$

$T[^\circ\text{C}]$	20	30	40	50	60	70	80	90	100	110	120
χ^2	1.224	1.229	1.233	1.236	1.237	1.237	1.237	1.235	1.229	1.210	1.151
$R_s [M\Omega]$	4.8	3.0	3.7	4.3	4.6	4.5	4.4	3.5	2.8	1.9	1.4
$C_g [nF]$	27.8	32.7	28.6	25.8	36.5	27.2	28.2	28.1	35.7	49.6	6.4
$R_g [\Omega]$	0.26	0.12	0.11	0.08	0.08	0.08	0.08	0.11	0.15	0.27	0.76
$f_g [MHz]$	22.0	40.6	50.6	77.1	54.5	73.2	70.6	51.5	29.7	11.9	32.7
$Y_o [pS-s^n]$	59.5	46.4	35.1	29.1	26.8	27.9	29.8	36.2	55.1	107.2	269.0
$n (x10^{-15})$	71.6	53.1	25.9	11.3	12.5	10.0	10.9	30.0	82.0	403.7	25940
$R_q [T\Omega]$	7.0	9.5	12.5	14.5	15.7	14.5	13.4	10.7	6.4	3.7	0.5
$C_{gb} [nF]$	81.7	73.2	72.0	68.3	52.5	63.4	63.2	74.0	74.6	92.5	82.9
$R_{gb} [G\Omega]$	575	669	826	972	1120	968	886	734	526	305	4
$f_{gb} [\mu Hz]$	3.39	3.25	2.68	2.40	2.71	2.59	2.84	2.93	4.06	5.64	480.20

Table 3Fitting parameters of the model $R(CR)(QR)(CR)$ of the Nyquist plot of Se→Sr NHFs varying the Se ion substitution ratios from 0.01 to 0.10 for $T = 20\text{ }^{\circ}\text{C}$.

$x[\%]$	0.01	0.02	0.03	0.04	0.05	0.06	0.08	0.10
χ^2	1.242	1.224	1.242	1.223	1.235	1.243	1.220	1.235
$R_s [M\Omega]$	78.2	4.8	11.8	3.2	2.3	15.1	3.5	5.8
$C_g [nF]$	0.59	27.8	20.4	31.7	25.5	20	35	29.4
$R_g [\Omega]$	0.06	0.26	0.03	0.27	0.15	0.03	0.27	0.16
$f_g [MHz]$	4500	22	260	18	41	265	17	34
$Y_o [pS-s^n]$	7.6	59.5	12.5	81.4	66.6	10.6	84.3	35.8
$n (x10^{-15})$	0.9	71.6	0.4	110.6	94.8	0.3	145.2	23.8
$R_q [T\Omega]$	20.7	7.0	25.3	4.4	3.6	26.6	4.2	11.2
$C_{gb} [nF]$	28.2	81.7	44.9	95.4	85.1	40.3	86.9	70.0
$R_{gb} [G\Omega]$	17	575	1396	363	318	1507	363	761
$f_{gb} [\mu Hz]$	332	3.4	2.5	4.6	5.9	2.6	5.0	3.0

boundaries increases from 22 MHz to 77 MHz up to 50 °C, then decreases from 73 MHz to 33 MHz, with a sharp drop to 55 MHz at 60 °C. The characteristic frequency for the grain fluctuates between 2.4 and 5.64 μHz up to 110 °C, followed by a sharp increase to 480.20 μHz at 120 °C. The frequency constant (n) for the constant phase element (CPE) at $x = 0.02$ fluctuates between 10 and 403×10^{-15} as temperature increases from 20 °C to 110 °C, with an exceptionally large value of $25,940 \times 10^{-15}$ at 120 °C. At 20 °C, n fluctuates between 0.3 and 145×10^{-15} as temperature increases up to 120 °C.

The Nyquist plot provides insight into the electrical behavior of Se-substituted Sr-NHFs. The characteristic frequencies of the grain and grain boundary are related to the material's conductivity and dielectric properties. The grain interior exhibits higher frequency responses (kHz to MHz range) due to faster charge transport, while grain boundaries show lower frequency responses (Hz to kHz range) due to higher resistance and capacitance. The characteristic frequencies are defined by:

$$f_g = 1/(2\pi R_g C_g) \text{ for grains} \quad (7a)$$

and

$$f_{gb} = 1/(2\pi R_{gb} C_{gb}) \text{ for grain boundaries} \quad (7b)$$

The Nyquist plot, represented by the real part ($\text{Re}Z$) versus the imaginary part ($\text{Im}Z$) of impedance, typically forms a semicircle or arc: firstly, a perfect semicircle indicates ideal Debye relaxation with a single time constant. Secondly, a depressed arc suggests non-Debye relaxation, indicating multiple time constants or distributed capacitances. Therefore, the high-frequency region corresponds to bulk grain properties, while the low-frequency region corresponds to grain boundary effects and electrode polarization. Se substitution modifies grain resistance by altering electronic or ionic transport mechanisms. Enhanced conductivity due to Se substitution shrinks the semicircle, indicating lower bulk resistance, whereas increased resistivity results in a larger semicircle. The substitution may introduce additional polarization mechanisms, affecting the curvature of the semicircle. Increased capacitance due to grain boundary or interfacial effects may shift the peak of $\text{Im}Z$ or broaden the semicircle.

Fitting the Nyquist plot to an equivalent circuit model helps analyze changes in impedance behavior. Increased frequency dispersion signifies complex relaxation processes influenced by Se substitution. If substitution improves charge transport, the arc radius decreases due to lower grain resistance. Multiple arcs may appear in the Nyquist plot due to additional charge carrier pathways or interfaces introduced by Se. Modelling the Nyquist plot using an equivalent circuit (e.g., R–C, R–CPE) as illustrated in Fig. 13 quantifies changes caused by Se substitution. Changes in grain and grain boundary resistances and capacitances reflect microstructural modifications. The presence of non-ideal capacitance, represented by a CPE, suggests heterogeneous capacitance distributions. A reduced arc radius at high frequencies indicates improved conductivity, whereas a second arc at low frequencies signifies increased grain boundary impedance or electrode polarization effects due to microstructural changes. Comparing pre- and post-substitution Nyquist

plots provides valuable insights into how Se influences the electrical and microstructural properties of Sr-NHFs.

4. Conclusion

Se→Sr NHFs were successfully prepared via sol-gel combustion route. The impact of Se on structure and morphology was investigated. The magnetic investigations of $M - H$ curves are consistent with the response of ferrimagnetic materials under an externally applied magnetic field. The registered $M - H$ hysteresis loops also indicated “wasp-waisted” behavior around $H = 0$ kOe for different prepared samples. M_s , M_r , and H_c were found to increase as the concentration of selenium ions increases, achieving their maximum values for the sample with $x = 0.05$, then reduced as the concentration of selenium ions further increased ($x > 0.05$). The maximum values achieved for the product with $x = 0.05$ might be useful in permanent magnet applications. It was also disclosed that the wasp-waisted $M - H$ loops and the large values of coercive fields for different prepared compositions suggest that particles consisted of both single-magnetic domains and multi-magnetic domains. Also, Selenium does not enter the crystal lattice and does not interact with the ions constituting the Ba–Sr M-type hexaferrite system, leading to the appearance of a secondary phase, which will greatly affect the magnetic properties. Comprehensive electrical and dielectric analyses demonstrated that Se substitution markedly affects conduction and polarization mechanisms. AC and DC conductivity results indicate that charge transport is dominated by a combination of hopping and thermally activated processes. The observed decrease in dielectric constant and dielectric loss with increasing frequency suggests prominent dipolar relaxation and interfacial polarization at lower frequencies. Cole–Cole plot analyses confirmed deviations from ideal Debye behaviour, revealing multiple relaxation processes, while Nyquist plot fittings highlighted distinct grain and grain boundary contributions modified by Se substitution. These findings underscore the tunable electrical and dielectric properties of Se-substituted Sr-NHFs, making them promising candidates for microwave absorbers, energy storage systems, and electronic components requiring controlled impedance and dielectric characteristics.

CRedit authorship contribution statement

Y. Slimani: Writing – original draft, Writing – review & editing, Visualization, Investigation, Formal analysis, Data curation. **B. Ünal:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis, Data curation. **A. Baykal:** Writing – review & editing, Writing – original draft, Validation, Supervision, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **M.A. Almessiere:** Writing – original draft, Writing – review & editing, Project administration, Methodology, Investigation, Formal analysis. **A. Demir Korkmaz:** Writing – original draft, Methodology, Investigation, Data curation. **S. Caliskan:** Software, Formal analysis, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.matchemphys.2025.131349>.

Data availability

Data will be made available on reasonable request.

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