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The effect of samarium substitution on the structure, conduction mechanism, and dielectric relaxation of MnNi-nanospinel ferrites

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The incorporation of Sm into Mn–Ni spinel ferrites to form $\text{Mn}_{0.5}\text{Ni}_{0.5}\text{Sm}_x\text{Fe}_{2-x}\text{O}_4$ ($x \leq 0.10$) nanospinel ferrites (Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSF) was achieved via the sol–gel method. The product was subjected to different characterization techniques to investigate its structure and morphology. A cubic crystal structure and a sphere morphology were confirmed by X-ray Powder Diffractometry (XRD), scanning electron microscope (SEM) and transmission electron microscope (TEM), respectively. The electrical and dielectric spectra of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSF were systematically examined using dielectric spectroscopy, electrical conductivity, and complex impedance analyses. The AC conductivity spectra reveal both frequency- and temperature-dependent dispersions, which are consistent with Jonscher's power law (JPL), whereas the Arrhenius analysis of the DC conductivity discloses two thermally activated regimes dominated by localized and long-range small polaron-hopping processes. The calculated activation energies ranged from 10 to 190 meV in the LT region and 420 to 530 meV in the HT region, confirming the dual conduction mechanism. Furthermore, dielectric constant and loss spectra demonstrated Maxwell–Wagner interfacial polarization and Koop's phenomenological behavior, with Sm^{3+} ion-substitution ratios most strongly influencing the grain boundary resistance and polarization dynamics. In addition, the complex electrical modulus and Nyquist plots reveal single semicircular arcs, indicating grain-boundary-dominant conduction and non-Debye relaxation. Interestingly, moderate Sm^{3+} ion-substitution ratios, ranging from 0.050 to 0.075, improve lattice distortion, lower the activation energy (E_A), and enhance hopping conduction; conversely, excessive substitution ratios, such as 0.10, lead to defect clustering, significantly reducing electrical performance. All these findings strongly highlight the critical role of rare earth (RE) element substitution in shaping conduction and dielectric relaxation in Mn–Ni NSF.

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1. Introduction

Spinel ferrites are mixed metal oxides that belong to the soft ferrite group. They hold significant technological promise due to their distinctive electrical, magnetic, and optical characteristics, such as low dielectric loss, high electrical resistivity, high magnetization, low eddy current losses, wide band gap, and unique electromagnetic properties. Spinel ferrites are valued

for electrical and dielectric applications, particularly in transformer cores, electronic filters, high-frequency power devices, electromagnetic shielding, and radar-absorbing systems.¹ Spinel ferrites are characterized by a spinel crystal structure and are generally expressed as AB_2O_4 (or MFe_2O_4). In spinel ferrites, the tetrahedral (Td, A) site is occupied by divalent metal cations (such as Mn^{2+} , Co^{2+} , Ni^{2+} , and Zn^{2+}), while the octahedral sites (Oh, B) are predominantly occupied by trivalent ferric ions (Fe^{3+}).² Spinel ferrites, known for their excellent physicochemical properties, adopt a closed cubic framework, with a cage-like lattice. Within this structure, the close packing of O^{2-} anions positioned at all corner lattice sites results in the formation of 96 interstitial void spaces. This arrangement provides a total of 64 Td (A) and 32 Oh (B) positions for cation distribution.³ However, cation occupancy is limited, and only 8 Td (A) and 16 Oh (B) sites are filled by cations. The relatively open framework of the flexible spinel lattice facilitates the migration of smaller cations from A- to B-sites and from B- to A-sites. The arrangement of cations between these two sites largely depends on their relative affinity, synthesis methods

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and conditions, ionic radii, interstice size, and stabilization energy.² Currently, complex spinel ferrites—formed by combining two or more single-component ferrites—are widely applied in transformer cores, inductor cores, telecommunication systems, and information-storage systems owing to their superior magnetic and electrical attributes.⁴ As in the case of spinel ferrites, spinel oxides (AB_2O_4) are important materials due to their electrical, mechanical, magnetic and optical properties, and they are suitable as superconductors, magnetic cores, and high-frequency devices. Zinc aluminate (ZnAl_2O_4), zinc gallate (ZnGa_2O_4) and zinc indate (ZnIn_2O_4) belong to this fascinating family of materials.^{5,6}

Among spinel ferrites, nickel ferrite (NiFe_2O_4) is notable for its superior magneto-electrical behavior, defined by high saturation magnetization, enhanced permeability, significant resistivity, elevated Curie temperature, and low eddy current dissipation. NiFe_2O_4 adopts an inverse spinel lattice, with Fe^{3+} ions equally occupying A and B sites, whereas Ni^{2+} ions occupy Oh (B) sites. This cationic arrangement gives rise to its ferromagnetic properties. Also, MnFe_2O_4 is a soft ferrite, exhibiting high magnetic permeability and low hysteresis loss, with Mn^{2+} ions residing at the Td (A) sites, whereas Fe^{3+} ions occupy Oh (B) sites. Ni^{2+} substitution in MnFe_2O_4 significantly modifies its structural, magnetic, electrical, and dielectric properties.⁷ Mixed Ni–Mn spinel ferrites are promising for soft and hard magnet applications due to their low dielectric losses, high resistivity, large MS, and high permeability, which have been studied by different authors.^{8–10} Doping spinel ferrites with rare-earth as well as transition metals has been shown to enhance their properties. Hulsure *et al.*¹¹ showed that samarium (Sm)-substituted $\text{Mn}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ spinel ferrite ($0.0 < x < 0.3$) maintained the cubic spinel structure, increased lattice parameters at higher concentration, and induced superparamagnetic behaviour. The highest saturation magnetization and magnetic moment were obtained at $x = 0.2$. These modifications strengthen

electrical transport and magnetic performance, supporting suitability for ferrofluid and heat-transfer applications. Taneja *et al.*¹² reported that Sm-substituted $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Bi}_{0.04}\text{Fe}_{1.96}\text{O}_4$ exhibited high DC resistivity (8.20×10^9 to $2.76 \times 10^{10} \Omega \text{ cm}$) and AC conductivity ranging from 1.80×10^{-5} to $5.48 \times 10^{-5} \text{ S cm}^{-1}$. The dielectric constant ($\epsilon' = 4.17\text{--}8.89$), dielectric loss ($\epsilon'' = 0.101\text{--}1.38$), and $\tan \delta$ ($0.069\text{--}0.133$ at 1 MHz) varied with the Sm concentration, indicating suitability for high-frequency systems, microwave applications and transformers. Siddiqui *et al.*¹³ reported that Sm-substituted spinel ferrite ($\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$) ($0.00 < x < 0.20$) exhibited a dielectric constant (ϵ') in the range of 2.17–6.92 and a dielectric loss (ϵ'') between 0.0667 and 3.34. Increasing the Sm content enhanced the ϵ' , ϵ'' , and AC conductivity (at frequencies above 1.6 GHz) up to an optimal concentration of $x = 0.15$, beyond which these properties showed a slight decline.

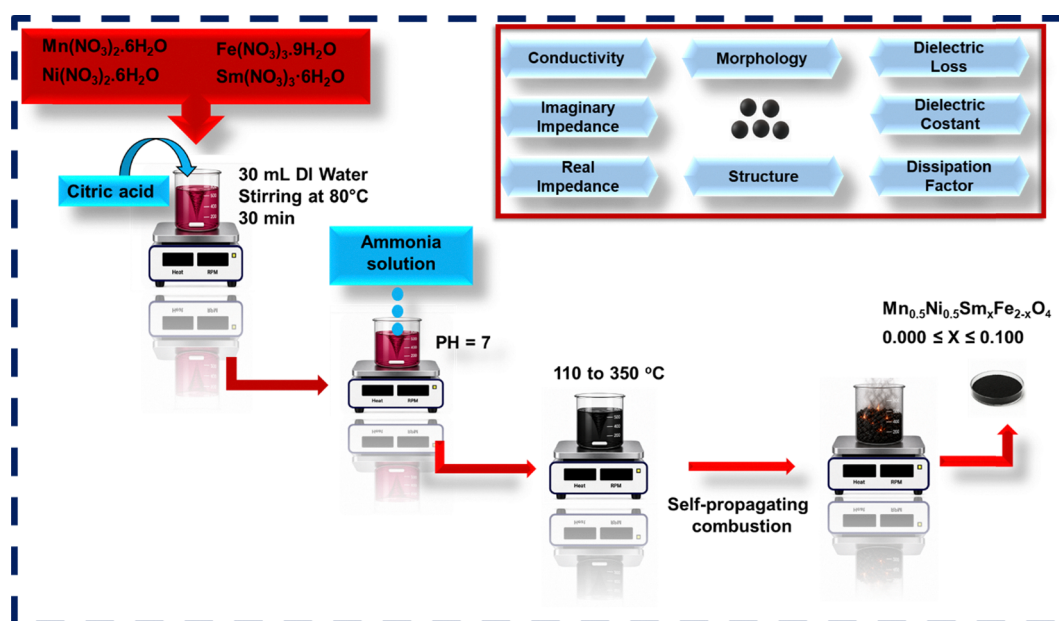
To date, studies investigating the role of Sm substitution in governing the conduction mechanism and dielectric relaxation of $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF are absent in the literature. Therefore, this study focused on investigating the effect of Sm doping ($x \leq 0.10$) in $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSFs with respect to their structural, electrical and dielectric properties.

2. Experimental

2.1. Chemicals and instrumentations

The following chemicals have been received from Merck ($\text{Mn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (99.9%), $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ($\leq 98.5\%$), $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ($\leq 98\%$), $\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ($\leq 98\%$) and citric acid ($\text{C}_6\text{O}_8\text{O}_7$) ($\leq 98\%$)) and used without further purification.

Phase identification and Rietveld refinement of XRD patterns were performed using Match 4 and FullProf software. Microstructure features and elemental composition analysis were examined by scanning electron microscopy (SEM, Tescan Vega 3), transmission



Scheme 1 Synthesis of the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSFs.

electron microscopy (TEM, Thermo Talos), and energy dispersive spectroscopy (EDS, AMETEK Element EDAX). Dielectric and electrical measurements of sintered ceramics were performed by dielectric impedance spectroscopy (Novocontrol Technologies) in a frequency range up to 1 MHz and a temperature range of 20–120 °C. Proton conductivity studies and dielectric measurements of the samples were performed using a Novocontrol impedance spectrometer. The films were sandwiched between gold blocking electrodes, and the conductivities were measured in the frequency range of 0.1 Hz–3 MHz at 10 °C intervals. The temperature was controlled by a Novocontrol cryosystem, which is applicable between 20 and 120 °C.

2.2. Synthesis

The Sm → MnNi ($x \leq 0.10$) NSF's were fabricated *via* the sol-gel method, with citric acid employed as fuel. The synthesis proceeded by dissolving a stoichiometric amount of metal nitrates and 3 g of citric acid in 30 mL of deionized H₂O (DI water) together under constant stirring at 80 °C for 30 min to get a homogeneous mixture. Then, a neutral pH (7) of the resultant mixture was attained by the addition of an ammonia solution, where the temperature increased from 110 to 350 °C, followed by constant stirring until the gel formed. Through the heating procedure, the resultant gel was burnt to get a black powder (self-propagating combustion). The obtained solid product was not subjected to any calcination process (Scheme 1).¹³

3. Results and discussion

3.1. Structure and morphology

The crystallite structure and phases of the Sm → MnNi ($x \leq 0.10$) NSF's were analyzed using an X-ray diffractometer (XRD). The XRD patterns shown in Fig. 1 disclose the prominent peaks of the pure phase of Mn–Ni cubic spinel ferrite. The spectrum shows the absence of the undesired Sm second phase, which indicates that the Mn–Ni spinel crystal is the dominant phase.^{13–15} The structural parameters, cell volume, XRD density, crystallite sizes (D_{XRD}) using the Debye–Scherrer equation and Rietveld reliability parameters were calculated using the Match4 software and are listed in Table 1. The structural parameters and crystal size exhibited a fluctuating increasing trend, then they decreased with increasing Sm ratios due to the variation in the distribution of ions from one ratio to another, as a result of a mismatch in ionic radii between the dopant and the host element. Fig. 2 reveals the SEM morphological analysis results for Sm → MnNi ($x \leq 0.10$) NSF's. According to the SEM analysis, all substituted samples show notable agglomeration of spherical particles of different diameters, forming a porous, sponge structure. Elemental analysis and mapping of the Sm → MnNi ($x = 0.050$ and 0.075) NSF's were determined *via* EDX (Fig. 3), revealing the presence of Mn, Ni, Sm, Fe and O. The TEM analysis showed a notable aggregates of small particles. The particle-size distribution histogram revealed that the average size was approximately 40 nm. The selected area electron diffraction pattern (SAED) and HR-TEM images exhibited the (111), (220), (311), (400) and (422) indexed planes, conforming to the

cubic spinel structure of the Sm → MnNi ($x = 0.075$) NSF's, as shown in Fig. 4.

The cation distribution of NSF's [(Sm → MnNi ($x \leq 0.10$))] was estimated by X-ray diffraction analysis employing the Bertaut structure-factor model. The Bertaut model considered the combined intensities of the (220), (400) and (440) spinel lines, which are known to be quite sensitive to variations in the distribution of the two Td and Oh sites. The site occupation analysis for $x = 0.000$ revealed that Mn resides in the A (Td) site, while Ni occupies the B (Oh) site, although iron may be accommodated in both the A and B sites. Furthermore, a constrained cation-distribution model was proposed, where Sm preferentially occupied the B site in all doped samples. Table 2 provides details of the cation distribution; the parentheses show the Td and Oh sites.^{16,17}

3.2. Electrical and dielectric properties

3.2.1. Scientific significance of Sm substitution. The significance of Sm³⁺ substitution is not limited to identifying an optimum composition. A quantitative comparison with previously reported rare-earth-substituted Mn–Ni, Mn–Zn, and Ni–Zn ferrites,^{4–18} considering crystallite size, activation energy, AC conductivity, dielectric response, and relaxation characteristics, indicates that Sm incorporation systematically modifies the electrical response through changes in structural disorder, interfacial polarization, and localized charge transport. The observed composition- and temperature-dependent evolution of conductivity and relaxation therefore provides insights into the relationship between Sm-induced structural modification and AC charge-transport mechanisms.^{11–13,18}

The dielectric and electrical characteristics observed in the present system may have relevance to high-frequency and energy-related applications; however, such implications should be regarded as potential rather than demonstrated applications. Direct assessment of application performance would require the measurement of parameters such as the dielectric loss at the intended operating frequency, breakdown strength, recoverable energy density, and cycling stability. Accordingly, the present study focuses on establishing the composition-, temperature-, and frequency-dependent physical mechanisms rather than making unverified device-performance claims.

3.2.2. AC conductivity spectra. A 2D AC conductivity ($\ln \sigma_{\text{AC}}$ vs. $\ln \omega$) analysis, depicted in Fig. 5, was conducted at temperatures ranging from 20 to 120 °C for the Sm → MnNi ($x \leq 0.10$) NSF's. The spectra demonstrate a strong coupling among frequency, temperature, and Sm concentration, indicating that charge transport involves both low-frequency interfacial polarization and high-frequency localized carrier hopping.

For the undoped sample ($x = 0.00$), a pronounced low-frequency plateau, followed by a rapid increase in conductivity at higher frequencies, is observed, consistent with quasi-dc transport dominated by resistive grain boundaries and subsequent localized hopping. Increasing temperature shifts the spectra upward, confirming thermally activated transport. At $x = 0.025$, a broad intermediate-frequency transition emerges, suggesting that Sm incorporation modifies the characteristic

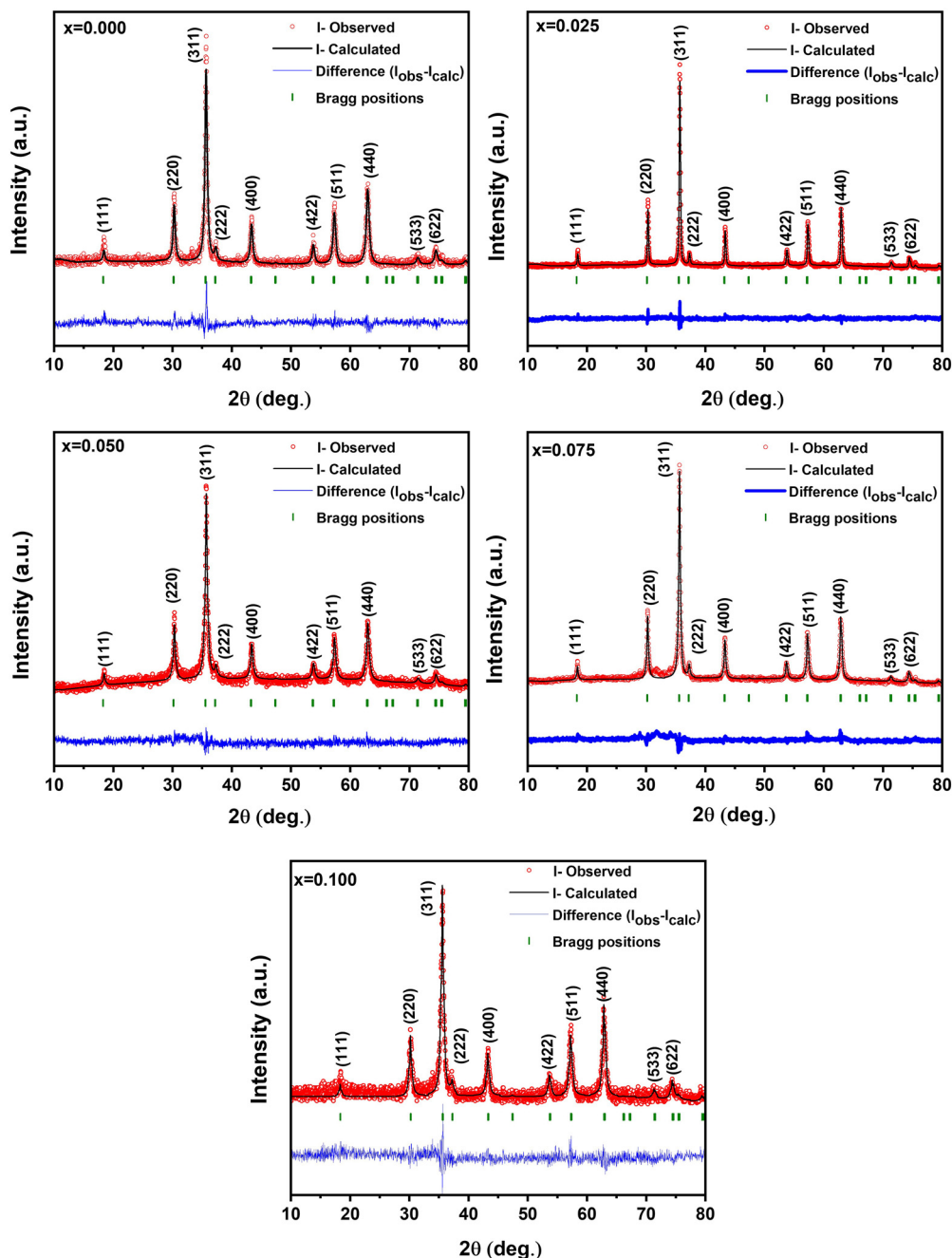


Fig. 1 Refined XRD powder patterns of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

relaxation times and redistributes the contributions of interfacial and intragranular transport. For $x = 0.050$, the conductivity

exhibits a smoother frequency dispersion, with temperature effects remaining strongest at low and intermediate frequencies.

Table 1 Refined structural parameters and Rietveld reliability parameters for the Sm $\rightarrow$ MnNi NSFs ($x \leq 0.10$) (lattice parameters (a_0), crystallite size (D_{XRD}), cell volume (V), X-ray density (d_x), Chi-square (χ^2) and Bragg R factor (R_{Bragg})

x	a_0 (Å)	V (Å ³)	d_x (g cm ⁻³)	D_{XRD} (nm)	R_p (%)	R_{wp} (%)	R_{exp} (%)	χ^2	R_{Bragg}
0.000	8.3495(4)	582.0260	5.264	36.3	34.3	38.2	38.4	0.992	12.8
0.025	8.3540(3)	583.0136	5.255	55.3	28.8	18.6	14.5	1.644	7.90
0.050	8.3486(3)	581.8879	5.265	50.5	47.5	33.1	24.9	1.777	24.3
0.075	8.3576(2)	583.7698	5.248	45.7	34.8	24.0	11.0	4.748	17.2
0.100	8.3491(5)	581.9967	5.264	54.5	47.6	49.4	43.6	1.282	33.1

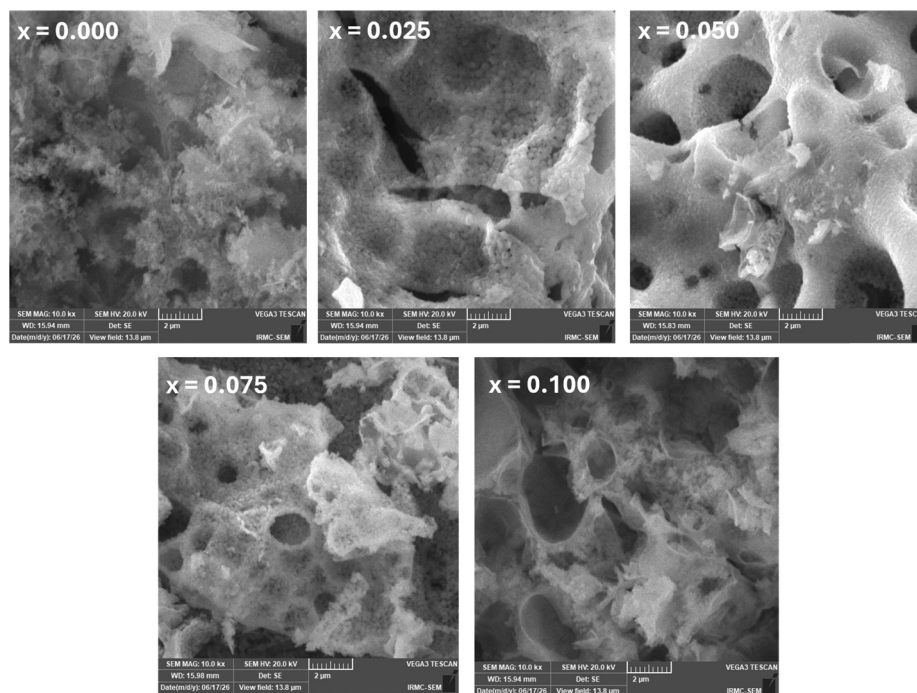


Fig. 2 SEM micrographs of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

This behavior indicates thermally assisted dispersive transport rather than a purely ohmic response. At $x = 0.075$, a distinct low-frequency plateau is again observed, whereas the spectra progressively converge at high frequencies. This convergence suggests that localized intragranular hopping becomes increasingly dominant as the influence of macroscopic grain-boundary barriers diminishes. The $x = 0.10$ composition displays a qualitatively different response, including a minimum/inflection region around $\ln \omega \approx 7-9$ and increased high-frequency fluctuations. These features suggest greater electrical heterogeneity and possible modification of localized states or interfacial barriers at high Sm concentrations, although they cannot, by themselves, establish a new conduction mechanism. Fundamentally, the systematic increase in σ_{AC} with temperature confirms thermally activated charge transport, likely involving localized hopping between mixed-valence cationic states.

The entire trend indicates that the conductivity is meaningfully dependent on both the applied angular frequency and the measured temperature, including substituted Sm^{3+} ion ratios, consistent with the typical behavior of NSFs. At lower frequencies, the σ_{AC} values remain relatively low and accordingly are almost temperature-independent. Such a tendency is due to the charge carrier accumulation at grain boundaries, acting as potential barriers to charge-carrier mobility. These relevant properties are consistent with Koop's phenomenological theory, which links the low-frequency response to the interfacial polarization between conductive grains and resistive grain boundaries.¹⁴

It is quite evident that as the frequency rises, the σ_{AC} parameter demonstrates a strong enhancement, particularly at higher temperatures. Such a variation in σ_{AC} suggests the

dominance of short-range hopping processes of localized charge carriers (*i.e.* small polarons) between Fe^{2+} and Fe^{3+} ions in the Oh (B) regions, where higher frequencies reduce the effect of grain boundary barriers. The notable transition from plane regions at low frequencies to dispersed regions at higher frequencies indicates a hopping conduction mechanism, as also reported for the Ni-Zn and Co-ferrites.^{14,15}

It should be noted that the increase in conductivity with temperature is consistent with enhanced mobility and/or hopping probability of localized charge carriers. The pronounced low-frequency dielectric dispersion is attributed primarily to the interfacial charge accumulation and Maxwell-Wagner-type polarization associated with electrically heterogeneous regions. Similarly, a significant temperature dependence is observed: σ_{AC} increases systematically with increasing temperature for all compositions. This thermally activated conductivity is most likely attributed to the increased probability of charge-carrier hopping with increasing lattice vibrations. At lower substitution levels in the range of 0.025–0.050, σ_{AC} increases more rapidly, which appears to be due to the facilitation of electron hopping due to local structural distortions arising from Sm^{3+} substitution. However, at higher substitution levels, such as $x = 0.10$, σ_{AC} is suppressed, likely due to the over-substitution reducing the $\text{Fe}^{2+}/\text{Fe}^{3+}$ ratio and thus limiting the available hopping sites.^{4,18,19} Furthermore, localized hopping between mixed-valence cations, particularly $\text{Fe}^{2+}/\text{Fe}^{3+}$ and possibly $\text{Ni}^{2+}/\text{Ni}^{3+}$, may contribute to the observed conductivity. However, these oxidation-state configurations are not directly established by the present electrical and dielectric measurements and are, therefore, considered plausible charge-transport pathways rather than definitive mechanisms. Oxygen-vacancy-related localized

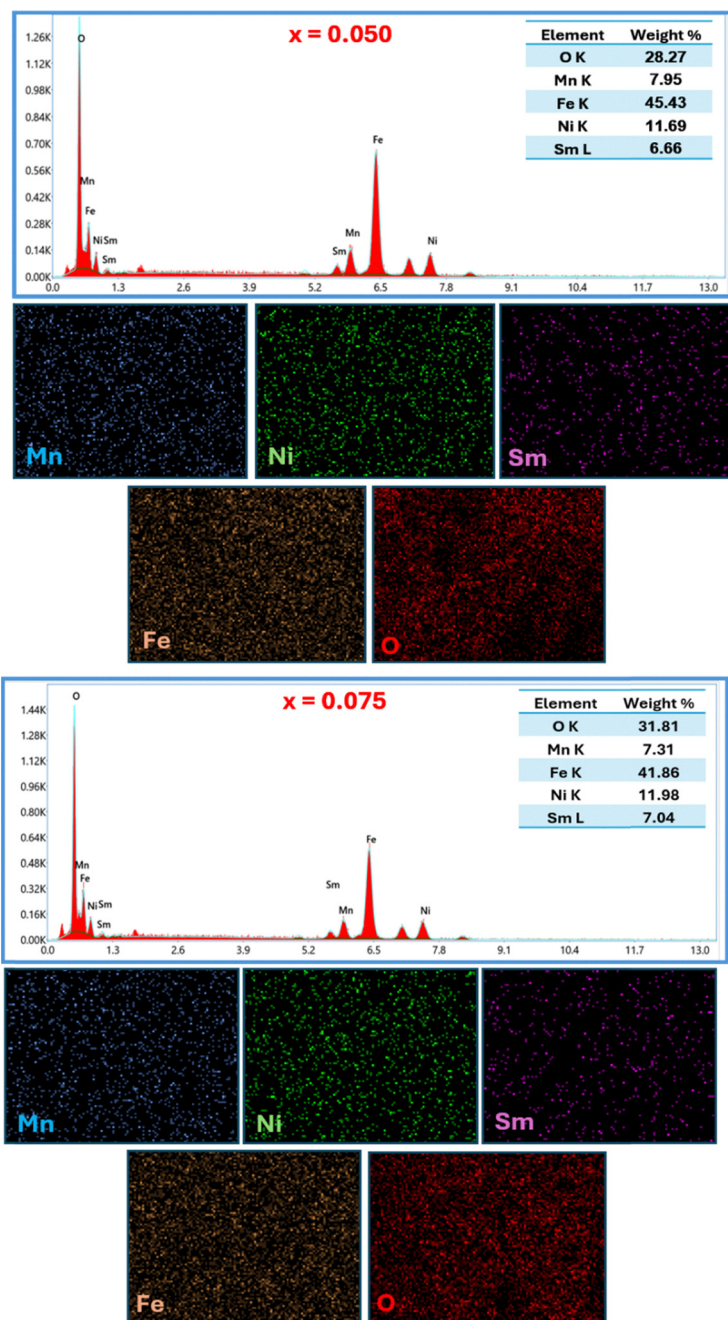


Fig. 3 EDX spectra and elemental mappings of the Sm $\rightarrow$ MnNi ($x = 0.050$ and 0.075) NSFs.

states may also contribute to charge trapping, polarization, and the frequency-dependent conductivity. The impedance and relaxation characteristics further support the thermally activated localized charge transport and electrically distinct regions within the NSFs.

As is known, the combined dependence on both frequency and temperature is an important finding confirming the following: the conduction mechanism in these ferrites is dominated by small polaron-hopping processes, the grain boundaries play a significant role at low frequencies, and intra-grain contributions are dominant at higher frequencies and temperatures. These

trends are consistent with the behavior reported in ferrites substituted with RE elements, where substitution generally reduces E_A at moderate levels but inhibits conduction at excessively high substitution rates.^{20–22}

3.2.3. Arrhenius analysis of the DC conductivity and activation energy (E_A). The Arrhenius analysis of the DC conductivity ($\ln \sigma_{DC}$ vs. $1/k_B T$) is presented in Fig. 6 for the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs. Accordingly, Fig. 6a exhibits the Arrhenius behavior of the DC conductivity for the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs. The linear dependence of $\ln \sigma_{dc}(x, T)$ on $1/k_B T$ over the investigated temperature range confirms thermally activated

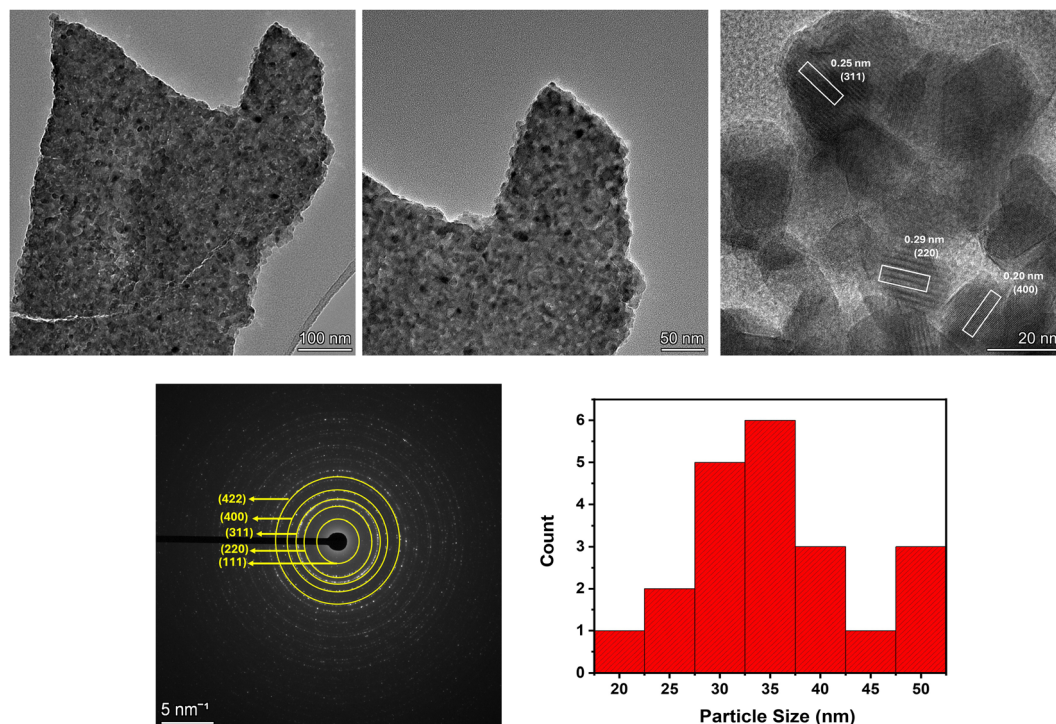


Fig. 4 TEM and HR-TEM micrographs, SAED pattern and particle-size distribution histogram of the Sm $\rightarrow$ MnNi ($x = 0.075$) NSFs.

Table 2 Cation distribution in the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs

x	Cation distribution
0.000	(Mn _{0.5} Fe _{0.5}) _A (Ni _{0.5} Fe _{1.5}) _B O ₄
0.025	(Mn _{0.5} Fe _{0.5}) _A (Ni _{0.5} Sm _{0.025} Fe _{1.475}) _B O ₄
0.050	(Mn _{0.5} Fe _{0.5}) _A (Ni _{0.5} Sm _{0.050} Fe _{1.45}) _B O ₄
0.075	(Mn _{0.5} Fe _{0.5}) _A (Ni _{0.5} Sm _{0.075} Fe _{1.425}) _B O ₄
0.100	(Mn _{0.5} Fe _{0.5}) _A (Ni _{0.5} Sm _{0.100} Fe _{1.4}) _B O ₄

charge transport. The extracted activation energy decreases from 465.8 meV for $x = 0.000$ to 431.8 meV for $x = 0.100$ (see Table 3), with intermediate compositions exhibiting nearly constant values of ~ 443 meV. The excellent linearity ($R^2 = 0.9967\text{--}0.9992$) supports the reliability of the Arrhenius analysis and indicates that Sm³⁺ ion incorporation progressively modifies the energetic barriers governing charge transport.

It is well known that the conductivity increases systematically with angular frequency, $\omega(2\pi f)$, confirming the contribution of localized charge-carrier hopping to AC transport. The frequency exponent $s(T, x)$, obtained from the $\ln \sigma - \ln \omega$ relationship, decreases monotonically from 0.631 at 20 °C to 0.323 at 120 °C (see Fig. 6b). This progressive reduction in $s(T, x)$ demonstrates the enhanced thermal activation of the charge carriers and a weakening of the frequency dependence of conductivity with increasing temperature. This behavior is characteristic of thermally assisted correlated hopping among localized charge states and is consistent with a barrier-controlled transport mechanism. The decrease in $s(T, x)$ suggests that increasing thermal energy facilitates carrier hopping over localized potential barriers, thereby increasing the

contribution of mobile carriers to the overall conductivity. Nevertheless, the declining R^2 values at elevated temperatures indicate that a simple power-law representation over the complete frequency range does not fully describe the data, emphasizing the importance of separating the frequency-independent σ_{DC} contribution from the dispersive $A(T, x)\omega^{s(T, x)}$ term. Consequently, quantitative fitting using the full Jonscher relation, together with $s(T)$ analysis, provides a more reliable basis for identifying the dominant hopping mechanism. It should be emphasized that the frequency exponent, $s(T, x)$, in Fig. 6b increases systematically with temperature for all compositions, indicating enhanced frequency-dependent localized charge dynamics at elevated temperatures. Notably, Sm³⁺ ion substitution strongly affects the magnitude of $s(T, x)$, where the highest values are obtained at $x = 0.100$, whereas comparatively low values are obtained at $x = 0.050$. This composition dependence suggests that Sm³⁺ ion incorporation modifies the distribution and mobility of localized charge carriers and associated hopping pathways.

Fig. 6c shows the corresponding temperature evolution of the Jonscher coefficient $A(T, x)$. The pronounced non-monotonic variations demonstrate that the strength of the dispersive AC-conduction contribution is strongly coupled to both temperature and Sm³⁺ ion concentration. Collectively, the systematic changes in $E_A(x)$, $s(T, x)$, and $A(T, x)$ indicate that Sm³⁺ ion substitution changes the balance between the thermally activated DC transport and the localized, frequency-dependent charge motion.^{14,23–25} These findings reinforce that Sm³⁺ ion substitution effectively influences the conduction mechanism in Mn–Ni NSFs and optimizes charge transport at moderate substitution levels.

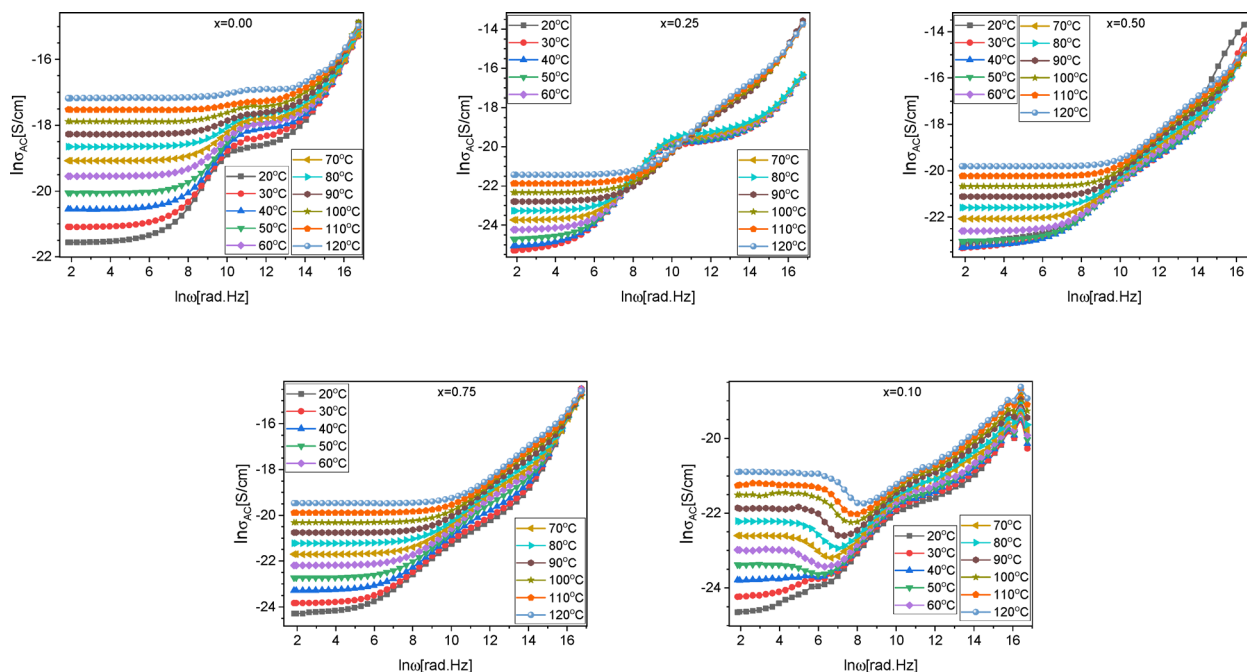


Fig. 5 AC conductivity spectra of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

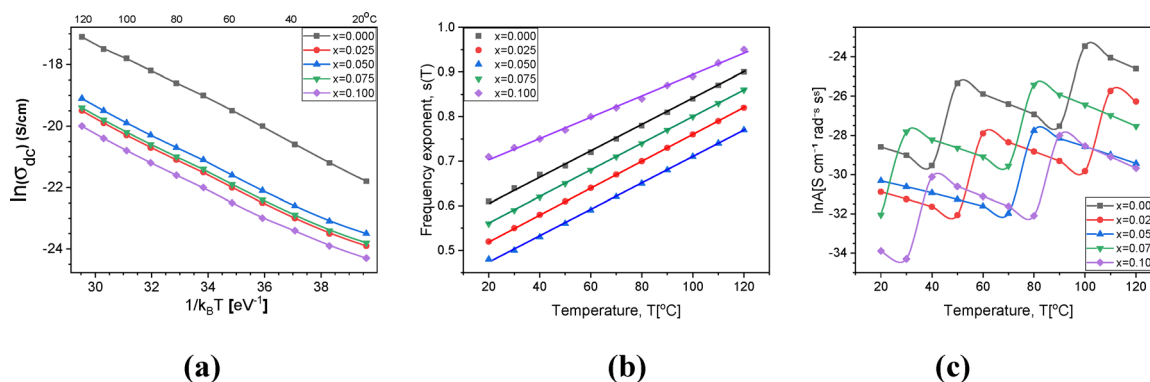


Fig. 6 (a) Arrhenius analysis of DC conductivity, (b) variation in the frequency exponent, $s(T,x)$, and (c) modified Bezier fittings of the Jonscher coefficient, $A(T,x)$, obtained from Jonscher's power law equation for the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

Table 3 E_A (in meV) of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs

x	E_A (meV)	R^2
0.000	465.8	0.9992
0.025	443.4	0.9978
0.050	443.5	0.9978
0.075	443.2	0.9978
0.100	431.8	0.9967

3.3. Dielectric constant spectra

The 2D characterization of the dielectric constant ($\ln \epsilon'$ vs. $\ln \omega$) of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs is presented in Fig. 7 as functions of frequency at temperatures ranging from 20 to 120 °C. The entire dielectric response exhibits pronounced frequency and temperature dependence, consistent with the

Maxwell-Wagner interfacial polarization model and Koop's phenomenological theory for heterogeneous dielectrics.^{26,27} It is clearly seen that at low frequencies, ϵ' reaches high values due to the accumulation of charge carriers at resistive grain boundaries, leading to strong space-charge polarization. Therefore, as the frequency increases, ϵ' decreases promptly and eventually reaches an almost constant value for the reason that the electric dipoles and localized charge carriers can no longer track the rapidly alternating electric field.^{14,15,28–30} It is noteworthy that this type of dispersion behavior confirms that the dielectric polarization is primarily governed by the Fe²⁺/Fe³⁺ and Ni²⁺/Ni³⁺ hopping processes in the Oh (B) regions.^{4,18,19,21,23–25}

Any enhancement in the ϵ' parameter with temperature is imperative, particularly at low and medium frequencies, which

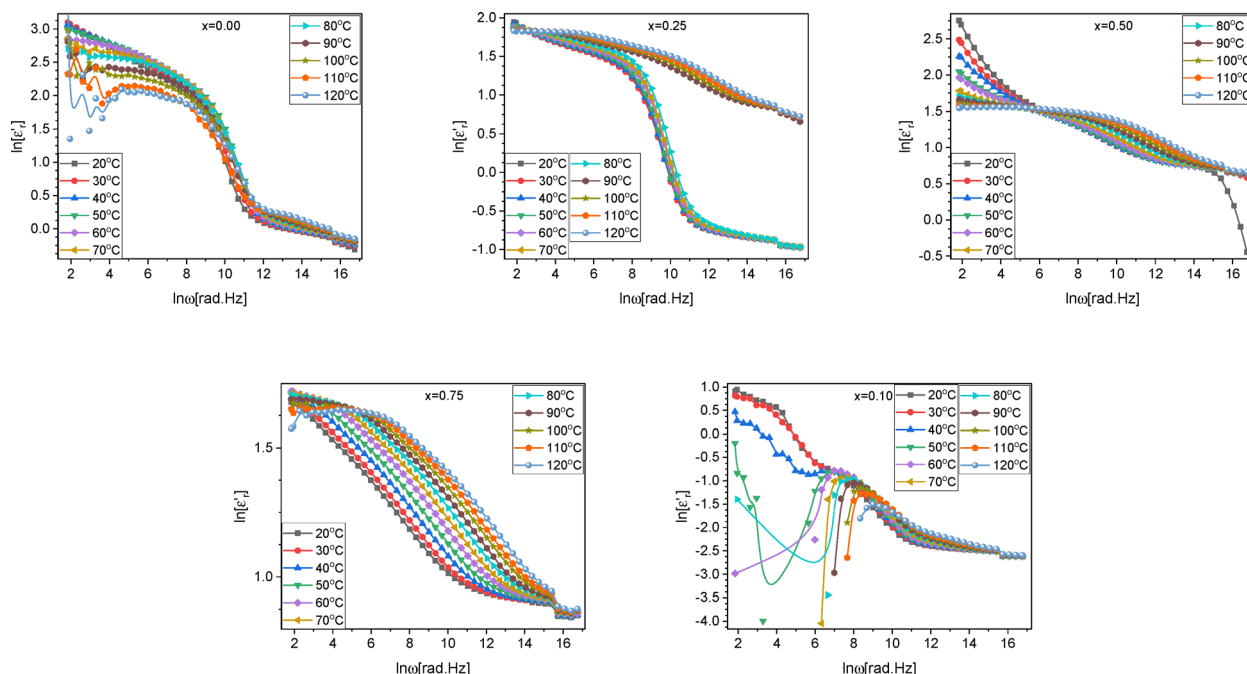


Fig. 7 Dielectric constant spectra of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

indicates thermally activated polarization resulting from improved charge-carrier mobility. Consequently, the Sm³⁺-ion substitution ratio strongly influences the dielectric response. It is noteworthy that medium substitution levels ranging from 0.025 to 0.075 yield smoother dielectric spectra and enhanced polarization stability, indicating optimized cation distribution and reduced defect clustering. Similarly, both the host NSF and highly substituted NSF exhibit more excessive fluctuations, representing cation disorder that negatively impacts structural

distortions, including dielectric polarization.^{26,27} Conclusively, the outcomes demonstrate that controlled Sm³⁺ ion incorporation effectively tunes the dielectric properties of MnNi NSFs, and moderate substitution provides the most stable dielectric response and improved polarization behavior suitable for high-frequency dielectric applications.²⁹

The enhanced dielectric permittivity observed at intermediate Sm concentrations, together with the associated relaxation behavior, indicates a potentially favorable dielectric response

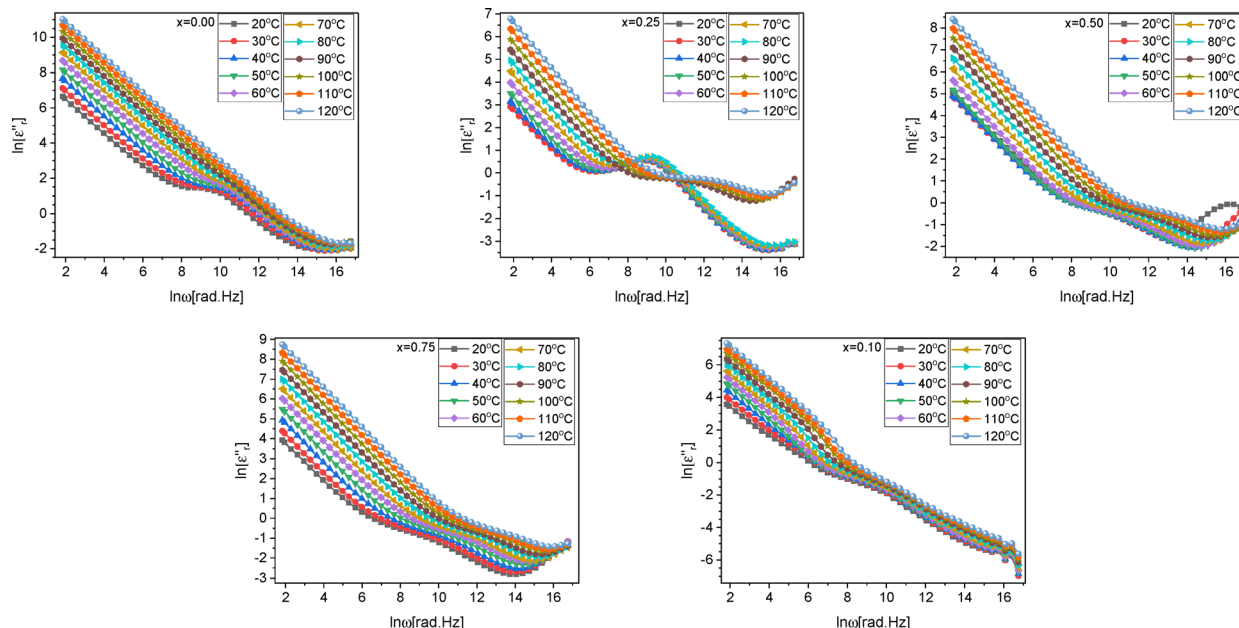


Fig. 8 Dielectric loss spectra of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

for energy-storage applications. However, the present dielectric and modulus measurements do not directly quantify the energy-storage performance. Definitive evaluation requires electric-field-dependent polarization (P - E) measurements and the determination of parameters such as recoverable energy density and energy-storage efficiency. Therefore, the observed dielectric enhancement is considered an indication of potential application relevance rather than direct evidence of improved energy-storage capacity.

3.4. Dielectric loss spectra

The 2D plots of the dielectric loss ($\ln(\epsilon'')$ vs. $\ln \omega$) are depicted in Fig. 8 as a function of the applied frequency and measured temperatures ranging from 20 to 120 °C for the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSF. Across all NSFs, the ϵ'' parameter exhibits strong dispersion with temperature and frequency, reflecting the competition between charge-carrier hopping transport and polarization processes. The ϵ'' behavior of the Sm $\rightarrow$ MnNi, ($x \leq 0.10$) NSFs is noteworthy for exhibiting a strong dependence on both frequency and temperature, revealing the relationship between charge-carrier hopping and polarization mechanisms, as mentioned before. It is very important to note that at low frequencies and high temperatures, ϵ'' is relatively high due to space-charge accumulation at grain boundaries, which is consistent with the Maxwell-Wagner interfacial polarization model and Koop's phenomenological theory.¹

It can be clearly observed that as the frequency increases, ϵ'' decreases, and a stable plateau is gradually reached as a result of the reduction in interfacial polarization and energy dissipation due to the inability of localized charge carriers ($\text{Fe}^{2+}/\text{Fe}^{3+}$ and $\text{Ni}^{2+}/\text{Ni}^{3+}$) to follow the rapidly alternating electric field, as mentioned earlier.^{23–25} Therefore, increasing the temperature increases the ϵ'' values through thermally activated small polaron-hopping processes. It is clearly seen that Sm³⁺ substitution significantly affects the dielectric response. Therefore, moderate Sm contents ranging from 0.025 to 0.075 suppress the ϵ'' value, stabilize the ϵ'' spectra, and promote structural homogeneity by limiting hopping conduction.^{26,27,29,31,32}

In contrast, the higher substitution ratio of 0.10 in NSF clearly leads to lattice disruption by disrupting the $\text{Fe}^{2+}/\text{Fe}^{3+}$ hopping paths, resulting in lower dielectric loss and reduced AC conductivity. As a result, the combined analysis of the dielectric constant, dielectric loss, and AC conductivity reveals a coherent conduction-relaxation mechanism governed by thermally activated small polaron hopping in the octagonal (B) regions. The above results are in good agreement with M-W polarization, Koop theory, and JPL, confirming that RE substitution effectively modifies the dielectric stability and electrical conduction properties of MnNi NSFs.^{14,15,33} It should be highlighted that the pronounced decrease in ϵ' and ϵ'' with increasing frequency, together with their enhancement at elevated temperatures and the observed relaxation features, is consistent with M-W interfacial polarization and Koop's phenomenological model. Accordingly, these trends indicate that localized charge carriers contribute more strongly at low frequencies, whereas their response becomes progressively limited at higher frequencies. The electric-modulus

response further supports localized relaxation processes. Thus, the M-W and Koop's models provide phenomenological frameworks consistent with the observed dielectric behavior, rather than uniquely established mechanisms.

3.5. Dissipation factor spectra

The dissipation factor, defined as the ratio of the imaginary and real components of dielectric permittivity, is a critical parameter that measures the energy dissipated within a dielectric material under a variable electric field, and it will provide comprehensive insights into the loss mechanisms in the dielectric system. Some 2D plots illustrating the dissipation factor or loss tangent ($\ln(\tan \delta)$, $\ln \omega$) for Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs with varying Sm³⁺ ion-substitution ratios are presented in Fig. 5 as a function of the angular frequency at temperatures up to 120 °C. A close examination reveals that the $\tan \delta$ spectra of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs are governed by a thermally activated relaxation process associated with M-W interfacial polarization and Koop's phenomenological model.

It is also evident that all NSFs exhibit distinct dielectric loss peaks, confirming relaxation attributable to electron hopping between ferrous and ferric ions in the Oh (B) sites. Consequently, it is noteworthy that as the temperature increases, the relaxation peaks systematically shift towards higher frequencies, indicating shorter relaxation times and, importantly, improved carrier mobility. This temperature dependency in the $\tan \delta$ loss parameter expresses Arrhenius-type kinetics and fascinatingly indicates that charge transport is governed by thermally activated small polaron hopping.

It is also worth noting that the Sm³⁺-ion substitution ratios significantly modify the dielectric loss features. Additionally, the host NSF exhibits a broad relaxation peak, indicating non-Debye relaxation, along with a dispersion of relaxation times resulting from structural heterogeneity. It is crucial to note that minor-to-moderate substitution ratios in the range of 0.025 to 0.075 modify the peak intensity and shape, indicating changes in charge-carrier concentration and grain boundary resistance without fundamentally affecting the conduction process, thereby preserving the underlying relaxation mechanism. Equally, a high Sm³⁺-ion substitution level of 0.100 in NSF produces numerous sharp relaxation peaks, reflecting the appearance of various relaxation mechanisms stemming from lattice disruption, defect formation, and changes in the local electronic ecosystem. It is important to note that these structural changes lead to multiple charge hopping pathways with different activation energies. Consequently, such results highlight a remarkable situation, showing that moderate Sm³⁺ ion incorporation preserves efficient dielectric relaxation, while excessive substitution leads to heterogeneous electrical behavior and complex relaxation dynamics, increasing dielectric losses and reducing dielectric stability (Fig. 9).²⁶

3.6. Real modulus (Re M) spectra

Some 2D plots depicting the real component of the electrical modulus (Re M vs. $\ln \omega$) as a function of frequency are presented in Fig. 10 at temperatures up to 120 °C from RT for the

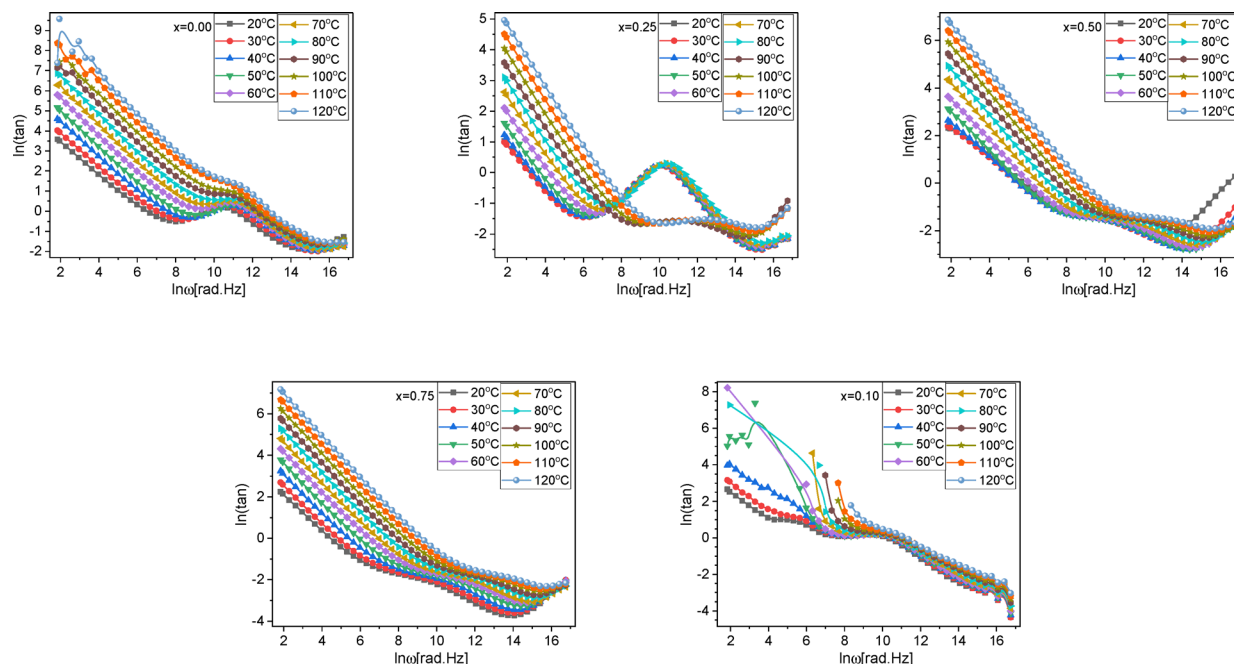


Fig. 9 Dissipation factor spectra of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs. ReM analysis is important for elucidating the dielectric properties of these NSFs because it provides information about their energy-storage capacities and relaxation dynamics. In addition, the modulus formalism effectively suppresses electrode polarization effects, which often conceal intrinsic bulk responses in the permittivity spectra.^{26,27} A close examination of the ReM spectra reveals the intrinsic dielectric relaxation behavior of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs. Unlike the complex permittivity spectral analysis, the modulus

formalism reduces the electrode polarization effects, allowing for a clearer assessment of grain-relaxation processes. Thus, it is a significant feature that all NSFs exhibit a characteristic sigmoid frequency dispersion, with predominantly long-range charge transport at lower frequencies, and the ReM parameter approaches zero because of electrode polarization. Furthermore, it is noteworthy that at high frequencies, the charge carriers rise to a plateau, unable to follow the externally applied electric field. It is also particularly important that the distribution region

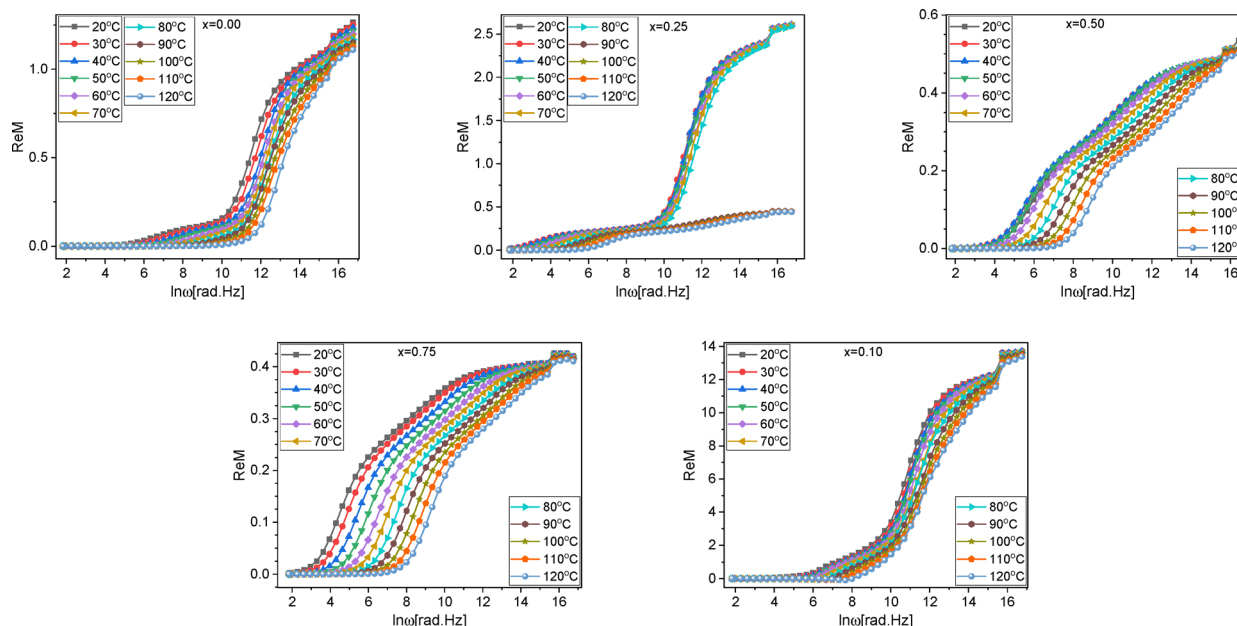


Fig. 10 Electrical real modulus spectra of the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

corresponds to the dielectric relaxation governed by short-range polaron-hopping processes.

The relaxation region shown in the given graphs shifts systematically towards higher frequencies with increasing temperature, validating thermally activated charge conduction and a decline in relaxation time, consistent with Arrhenius treatment. Therefore, it is observed that the Sm^{3+} ion-substitution level significantly affects the dielectric response in a nonlinear manner. A slight substitution ratio of 0.025 indicates a decrease in high-frequency permittivity, attributed to an increase in the $\text{Re}M$ value. However, moderate substitution ratios, such as 0.050 and 0.075, show improved dielectric polarization and increased energy-storage capacity, attributed to a decrease in the $\text{Re}M$ value.

Similarly, a high Sm^{3+} ion-substitution ratio of 0.10 results in a significant boost in the $\text{Re}M$ amount, corresponding to a substantial suppression of dielectric polarization, attributable to lattice disruption, defect formation, and disruption of ferrous-ferri hopping pathways. Consequently, $\text{Re}M$ analysis demonstrates that moderate Sm^{3+} ion incorporation governs both dielectric relaxation processes and energy-storage capacities, while an excessively high substitution level deteriorates dielectric performance through structural and charge-transport limitations.

3.7. Imaginary modulus ($\text{Im}M$) spectra

The relaxation behavior was further evaluated from the maxima of the imaginary modulus ($\text{Im}M$) and impedance ($\text{Im}Z$) spectra. The characteristic relaxation frequency (f_{max}) was determined from these maxima, and the corresponding relaxation time was calculated as ($\tau = 1/(2\pi f_{\text{max}})$). The temperature-dependent shift of the relaxation maxima toward higher frequencies, accompanied by a decrease in τ , confirms the thermally activated nature of the

relaxation process. The normalized $\text{Im}M$ and $\text{Im}Z$ spectra were also examined to assess the scaling behavior. The absence of complete spectral superposition, together with composition-dependent broadening, asymmetry, and occasional shoulder-like features, indicates a distribution of relaxation times. These features suggest that the observed relaxation involves overlapping bulk/grain, grain-boundary, and interfacial contributions rather than a single ideal Debye relaxation process.

Normalized imaginary components ($\text{Im}M$) of the complex electrical modulus are depicted in Fig. 11 as a function of frequency at temperatures ranging from RT up to 120 °C for the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSFs. Thus, the $\text{Im}M$ spectra serve as a powerful tool for studying dielectric relaxation because the observed top portions in the plots directly correspond to relaxation processes and are predominantly sensitive to grain contributions.

The normalized modulus spectra show a clear temperature-dependent displacement of the relaxation peaks toward higher frequencies with increasing temperature. This corresponds to a progressive reduction in the characteristic relaxation time, $\tau_M = (2\pi f_{\text{max}, \text{Im}M})^{-1}$, which is also evident in the relaxation-time plot. Thus, the relaxation process is thermally activated.

For $x = 0.000$, the normalized $\text{Im}M$ curves exhibit a relatively well-defined principal relaxation peak, although the low- and high-frequency sides are not perfectly symmetric. The deviation from an ideal symmetric Debye profile indicates a distribution of relaxation times rather than a single relaxation time. The behavior becomes considerably more complex for the intermediate compositions, particularly $x = 0.025$ and $x = 0.050$. Their normalized curves exhibit broadening, shoulders and, in some temperature ranges, more than one characteristic feature. This behavior is important in the context of the referee's

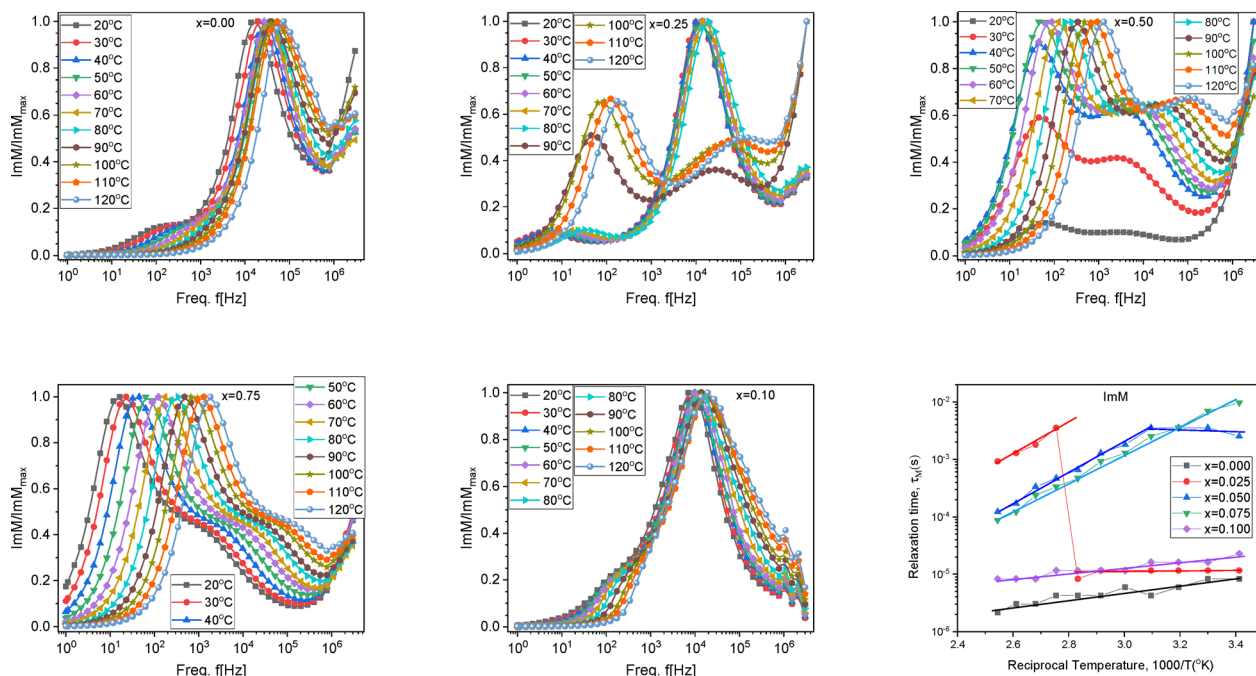


Fig. 11 Normalised imaginary modulus ($\text{Im}M$) spectra and the relaxation-time dispersion for the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSFs.

comment because it suggests that the measured response cannot necessarily be represented by a single Debye relaxation. The coexistence of bulk/grain and grain-boundary contributions can produce overlapping relaxation processes. For $x = 0.075$, the peaks shift systematically toward higher frequencies as temperature increases, while the peak profiles remain relatively broad. This again supports the thermally activated distributed relaxation. The $x = 0.100$ sample shows comparatively well-defined and nearly symmetric normalized peaks over much of the investigated temperature range, although some broadening is still visible on the high-frequency side. The normalized $\text{Im } M$ results provide much stronger evidence for non-Debye behavior than simply stating that the peaks are broad.

It is significant that the $\text{Im } M$ spectra of the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF s reveal that the dielectric relaxation is dominated by a thermally activated polaron-hopping mechanism. In all NCs, $\text{Im } M$ exhibits a distinct relaxation peak that systematically shifts towards higher frequencies with increasing temperature, validating the decline in the relaxation time and Arrhenius-type transport. Therefore, the broad relaxation peaks indicate non-Debye behavior, resulting from a dispersion of relaxation times relevant to structural heterogeneity. It is noteworthy that Sm substitution exerts a pronounced, non-monotonic effect on the relaxation dynamics. Therefore, both host ($x = 0.00$) and slightly substituted ($x = 0.025$) NCs exhibit well-defined relaxation peaks; the increasing peak intensity at $x = 0.025$ indicates improved dipole activity.

In contrast, intermediate NCs in the range of 0.050–0.075 exhibit suppressed relaxation peaks with strong low-frequency conductivity contribution, suggesting enhanced charge transport masking dielectric relaxation. At the highest Sm^{3+} ion concentration, $x = 0.10$, a sharp and intense relaxation peak reappears; this reflects a more insulating response that allows for decreased σ_{DC} and clear resolution of the intrinsic dielectric relaxation.

The combination of $\text{Re } M$, $\text{Im } M$, and $\tan \delta$ analyses consistently exhibits that dielectric relaxation and electrical conduction originate from small polaron-hopping processes in ferrous/ferric ion sites, which is a highly reasonable deduction. Additionally, moderate Sm^{3+} ion substitution enhances dielectric polarization, relaxation performance, and energy-storage capacity, even though excessive substitution at $x = 0.10$ leads to lattice distortion and defect-induced degradation of hopping transport, resulting in stunned polarization and degraded dielectric performance.

3.8. Complex electric modulus: Nyquist ($\text{Im } M$ vs. $\text{Re } M$) analysis

Fig. 10 presents the Nyquist plots of the complex electrical modulus ($-\text{Im } M$ vs. $\text{Re } M$) for the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF s measured between 20 °C and 120 °C. It is well known that the electric modulus formalism ($M^* = 1/\epsilon^* = \text{Re } M + j \text{Im } M$) is a useful approach in the analysis of dielectric properties because it suppresses electrode polarization effects and highlights grain contributions that are often hidden in impedance spectra by grain boundary processes. In each composition, each corresponding

graph shows semicircular arcs, representing different measurement temperatures.^{1,15,21,23,24,34–36}

Nyquist plots of the complex modulus ($-\text{Im } M$ vs. $\text{Re } M$) show that the dielectric response of the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF s is governed by grain-controlled, thermally activated relaxation processes. Thus, the complex modulus formalism effectively suppresses electrode polarization, allowing for the investigation of intrinsic grain contributions. All NSF s exhibit depressed semicircular arcs, demonstrating non-Debye relaxation associated with the dispersion of relaxation times, resulting from structural disorder and compositional inhomogeneity. Thus, the high-frequency intersection corresponds to intrinsic modulation ($M^\infty = 1/\epsilon^\infty$), while the occasional low-frequency spiky lines, especially for $x = 0.050$, designate residual long-range transport or minor electrode polarization effects.^{1,15,18,23,33–38}

As can be clearly seen from the relevant graphs, increasing temperature reduces the semicircular arc diameter and shifts the relaxation towards higher frequencies, resulting in shorter relaxation times and an increase in small-ferrous/ferric polaron hopping, consistent with Arrhenius-type transport. Therefore, it is clear that Sm substitution has a significant non-monotonic effect on the dielectric properties. Furthermore, while the host ($x = 0.00$) and lightly substituted ($x = 0.025$) NCs exhibit well-defined relaxation arcs, intermediate compositions ($x = 0.050$ – 0.075) present significantly lower modulus values, reflecting improved dielectric permittivity and electrical conductivity.

In contrast to all of the above, the high substitution ratio ($x = 0.10$) results in exceptionally high modulus values ($\text{Re } M \approx 14$, $\text{Im } M \approx 4.5$), indicating a low dielectric constant, suppressed conductivity, and a highly insulating condition stemming from lattice distortion and the disruption of the Fe^{2+} – Fe^{3+} hopping network. These results demonstrate that controlled Sm^{3+} ion incorporation effectively shapes the electro-dielectric performance of MnNi –SFs (Fig. 12).^{1,15,21–23,33–35}

3.9. Real impedance ($\text{Re } Z$) analysis

The real impedance, $\text{Re } Z$, depiction as a function of frequency for $\text{Sm} \rightarrow \text{MnNi}$ NSF s with varying Sm^{3+} substitution levels ($x = 0.00, 0.025, 0.050, 0.075$, and 0.10) over the temperature range of 20–120 °C is illustrated in Fig. 13. The $\text{Re } Z(f, T)$ spectra of the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF s reveal the evolution of charge-transport mechanisms with Sm^{3+} substitution. It is noteworthy that at low frequencies, all NSF s exhibit high $\text{Re } Z$ values, owing to the dominant contribution of grain boundaries, where space charge accumulation hinders charge transport. Therefore, as the frequency increases, $\text{Re } Z$ decreases significantly, indicating the suppression of interfacial polarization and the increase in electron hopping between ferrous and ferric ions in the Oh (B) regions. Accordingly, this finding highlights the primary conduction mechanism within the grains.^{4,5,10–12}

It is worth noting that the Sm^{3+} -ion incorporation significantly affects the impedance response. For the host NSF ($x = 0.00$), a gradual drop in the $\text{Re } Z$ intensity with increasing frequency and temperature is observed, consistent with thermally activated hopping conduction. The more pronounced decline in the $\text{Re } Z$ intensity at mid-frequencies for $x = 0.025$

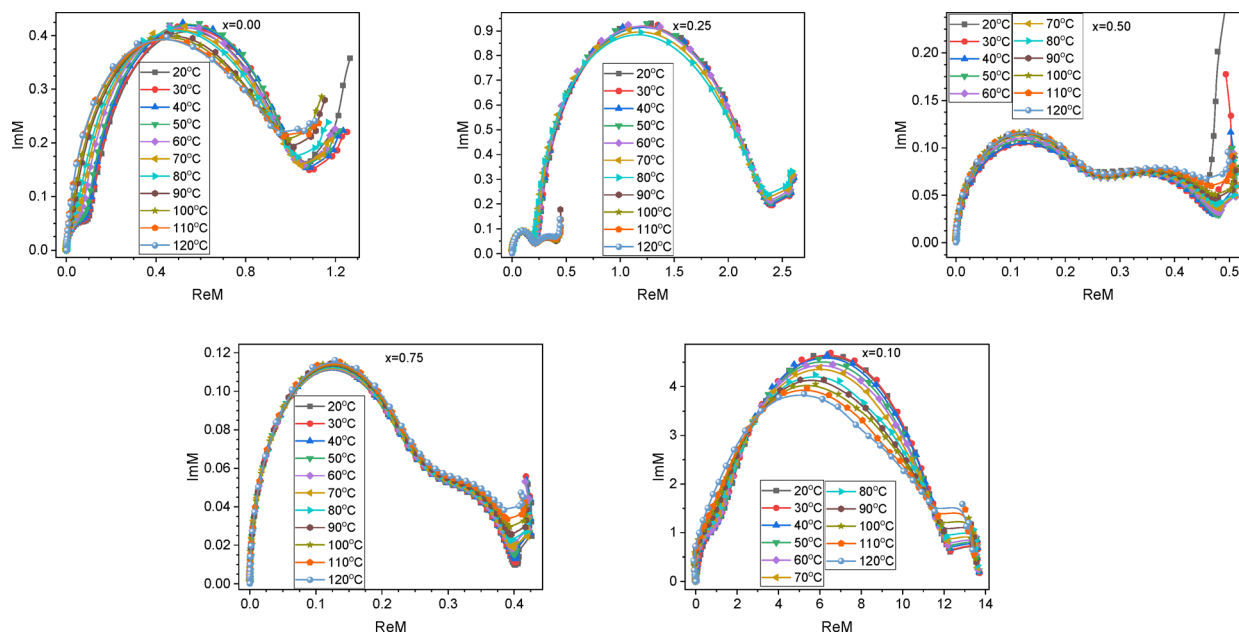


Fig. 12 Nyquist spectra of the complex electrical modulus ($\text{Im}M$ vs. $\text{Re}M$) of the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSFs.

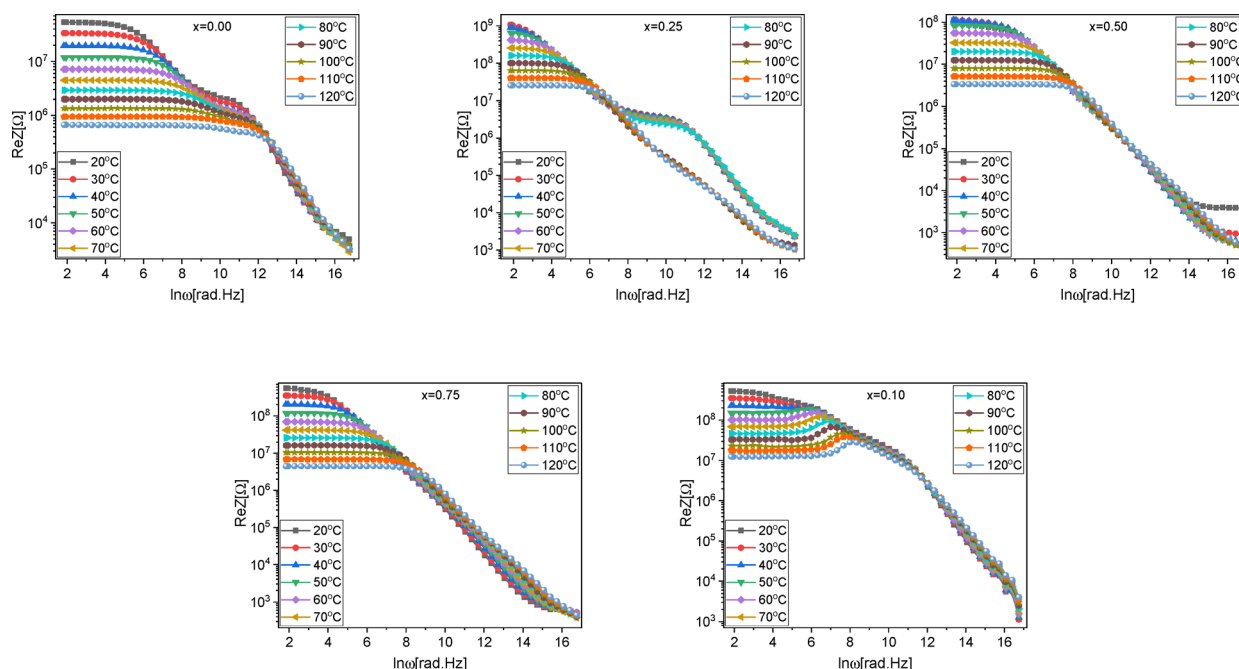


Fig. 13 Real impedance ($\text{Re}Z$) spectra of the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSFs.

is quite significant, indicating that Sm^{3+} ion-induced defect states support localized carrier hopping. NSFs with both $x = 0.050$ and 0.075 values exhibit the lowest impedance values and the smoothest frequency dependence, suggesting an optimal stability between lattice uniformity and defect influence that facilitates charge transfer. Conversely, an excessive Sm^{3+} ion-substitution level of 0.10 leads to high impedance at low frequencies due to structural disorder, defect

clustering, and carrier confinement, hindering hopping transport.^{22–24,35–38}

Furthermore, the systematic drop in the $\text{Re}Z$ intensity with increasing temperature for all NSFs is noteworthy, confirming the thermally activated small polaron-hopping processes. Expressly, all these results indicate a shift from grain boundary-dominant conduction at lower frequencies to intra-grain conduction at higher frequencies, with moderate Sm^{3+}

ion substitution effectively enhancing the electrical conduction mechanism.^{18,19,22,23,35–38}

3.10. Imaginary impedance ($-\text{Im} Z$) analysis

Normalized imaginary components ($-\text{Im} Z$) of the complex impedance are depicted in Fig. 14 as a function of frequency at temperatures ranging from RT up to 120 °C for the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSF. For all compositions, increasing temperature generally shifts the $\text{Im} Z$ relaxation maximum toward higher frequencies. According to $\tau_Z = (2\pi f_{\text{max,Im} Z})^{-1}$, this shift corresponds to a decrease in the relaxation time with temperature. The relaxation-time plot in Fig. 14 confirms this trend particularly clearly for $x = 0.025, 0.050, 0.075$ and 0.100 .

The $x = 0.025, 0.050$ and 0.075 compositions display relatively broad $\text{Im} Z$ peaks, demonstrating that the relaxation is not characterized by a single sharply defined time constant. The broadening is consistent with a heterogeneous electrical microstructure containing regions with different relaxation characteristics. In contrast, the normalized $\text{Im} Z$ spectra for some compositions, particularly $x = 0.025$, show a relatively smooth and systematic peak displacement with temperature. This suggests a dominant relaxation mechanism in these samples. The $x = 0.100$ spectrum is more complicated at lower temperatures, where overlapping features are apparent. This may indicate that more than one electrical relaxation contributes to the impedance response. The normalized $\text{Im} Z$ representation therefore supports the existence of distributed and/or overlapping relaxation processes, rather than a purely ideal Debye response.^{1,14,18,19}

It should be noted that the imaginary impedance ($-\text{Im} Z$) depiction delivers important information regarding the dielectric relaxation and conduction mechanisms of the ecosystem. In all NSFs, a distinct relaxation peak is observed in the $-\text{Im} Z$ spectra as functions of frequency and temperature, confirming

a thermally activated relaxation process associated with charge-carrier hopping between ferrous and ferric ions in the Oh (B) sites. Of particular importance is the finding that all NSFs exhibit some distinct relaxation peaks that systematically shift toward higher frequencies with increasing temperature, indicating both shorter relaxation times and improved charge-carrier mobility. Subsequently, the broad relaxation peaks observed in the host NSF disclose non-Debye behavior, resulting from the dispersion of relaxation times associated with structural inhomogeneity.^{21–24,34–38}

It is imperative to underline that the Sm³⁺ ion-substitution ratios significantly vary the relaxation dynamics. For a light substitution ratio of $x = 0.025$, slightly sharper maxima are generated, reflecting improved lattice homogeneity and reduced structural disorder. Besides, the intermediate substitution levels in the range of $x = 0.050$ – 0.075 produce well-defined, symmetrical relaxation peaks, reflecting an optimized local electronic structure, reduced defect scattering, and improved ferrous/ferric hopping transport. In contrast, a heavy substitution ratio of 0.10 broadens the relaxation peaks and weakens temperature dependencies, indicating that lattice disruption, defect clustering, and carrier trapping hinder long-range charge transport and broaden the relaxation time distribution, as mentioned earlier.^{18,19,22,37,38}

Therefore, it can be concluded that the systematic decrease in $-\text{Im} Z$ with increasing temperature further demonstrates the inverse relationship between impedance and conductivity, confirming thermally activated conduction. Overall, the consequences prove that moderate Sm-ion involvement optimizes dielectric relaxation and electrical conduction, while excessive substitution degenerates the electrical response by disrupting the hopping processes and increasing structural disorder.^{1,31,34–38}

For the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs, a reasonable physical interpretation is as follows: “electrode/interfacial polarization” +

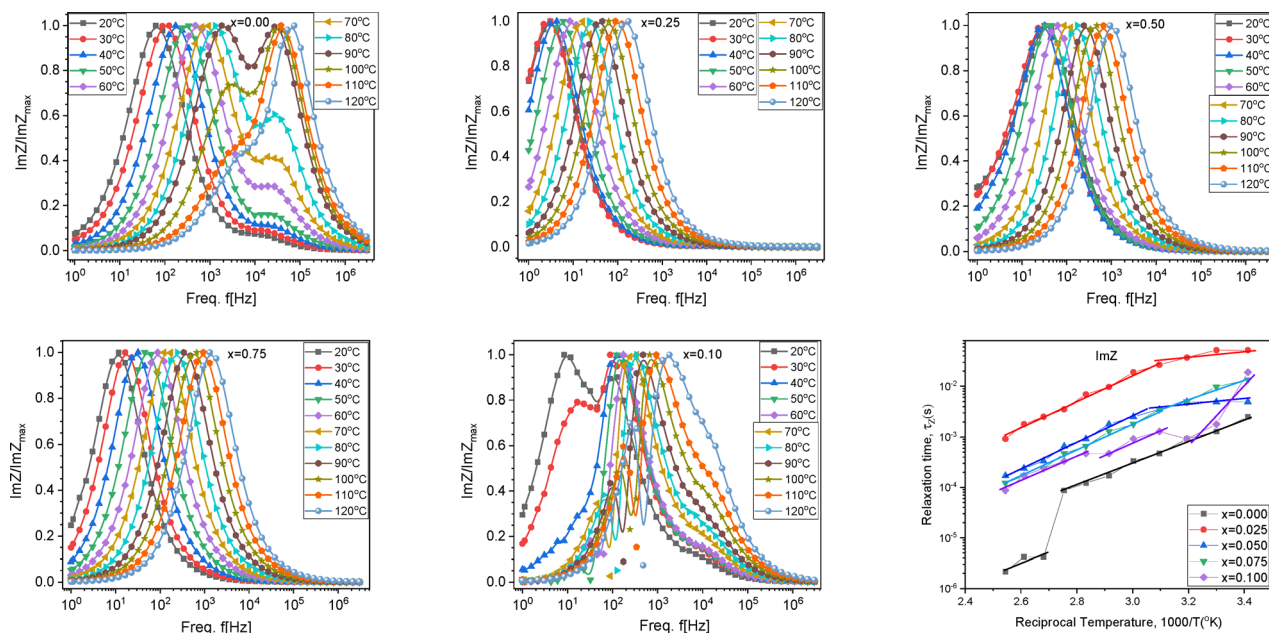


Fig. 14 Normalised imaginary impedance ($-\text{Im} Z$) spectra and the relaxation time dispersion for the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs.

“grain relaxation” + “grain boundary relaxation,” with the relative contribution changing with temperature and Sm concentration. Therefore, when $\text{Im}M$ and $\text{Im}Z$ show different peak shapes or characteristic frequencies, this can be interpreted as evidence that more than one electrically active relaxation process contributes to the overall response. The absence of complete superposition of the normalized $\text{Im}M$ and $\text{Im}Z$ spectra indicates that the relaxation-time distribution is affected by temperature and the Sm concentration, consistent with a non-Debye relaxation response and the coexistence of overlapping electrical relaxation processes.

The impedance response was interpreted by distinguishing the possible contributions of the grain, grain boundary, and electrode/interfacial regions. A depressed semicircle in the Nyquist plot alone was not considered sufficient evidence for grain-boundary-dominated conduction. Therefore, the impedance spectra were quantitatively analyzed using an equivalent-circuit approach incorporating resistive, capacitive, and constant-phase-element (CPE) components to account for non-ideal relaxation behavior. The extracted resistance and capacitance/CPE parameters, their temperature dependence, and the fitting quality were used as complementary evidence for identifying the electrically active regions. In contrast, the modulus representation was primarily used to evaluate localized relaxation and bulk/grain response, rather than to independently assign the observed relaxation to grain boundaries. Accordingly, the conduction-pathway interpretation is based on the combined quantitative impedance and modulus analyses rather than on qualitative assignment of individual spectral features.

The normalized $\text{Im}M$ and $\text{Im}Z$ spectra provide quantitative evidence of distributed electrical relaxation in the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF. For all compositions, increasing temperature progressively shifts the relaxation maxima toward higher

frequencies, corresponding to a reduction in the characteristic relaxation time, $\tau = (2\pi f_{\text{max}})^{-1}$. This thermally induced frequency displacement indicates thermally activated charge-relaxation dynamics. The normalized spectra, however, do not exhibit complete superposition, and several compositions show pronounced peak broadening, asymmetry and shoulders, particularly at intermediate Sm-substitution ratios. These features indicate a distribution of relaxation times rather than an ideal Debye process and suggest the coexistence of overlapping electrical responses associated with the heterogeneous grain/grain-boundary microstructure. The $\text{Im}M$ and $\text{Im}Z$ representations provide complementary information: the modulus response emphasizes bulk relaxation, whereas the impedance response is more sensitive to resistive microstructural contributions. The differences between their characteristic relaxation frequencies and spectral shapes therefore suggest that the measured response involves more than one relaxation process. The systematic decrease in τ with increasing temperature further supports thermally activated relaxation. Nevertheless, definitive identification of the relaxation-time distribution requires quantitative Arrhenius fitting of $\ln \tau$ versus $1/T$ and the extraction of a Cole-Cole or Havriliak-Negami distribution parameter.

3.11. Complex impedance: Nyquist ($-\text{Im}Z$ vs. $\text{Re}Z$) analysis

In Fig. 15, Nyquist plots of all the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF are shown for some depressed semicircular arcs whose diameters progressively decrease with increasing temperature, which demonstrates the negative temperature resistance coefficient (NTCR), characteristic of semiconductor ferrites and enhanced charge carrier mobility. This behavior imitates the thermally activated nature of conduction, as higher temperatures facilitate electron hopping between ferrous and ferric ions

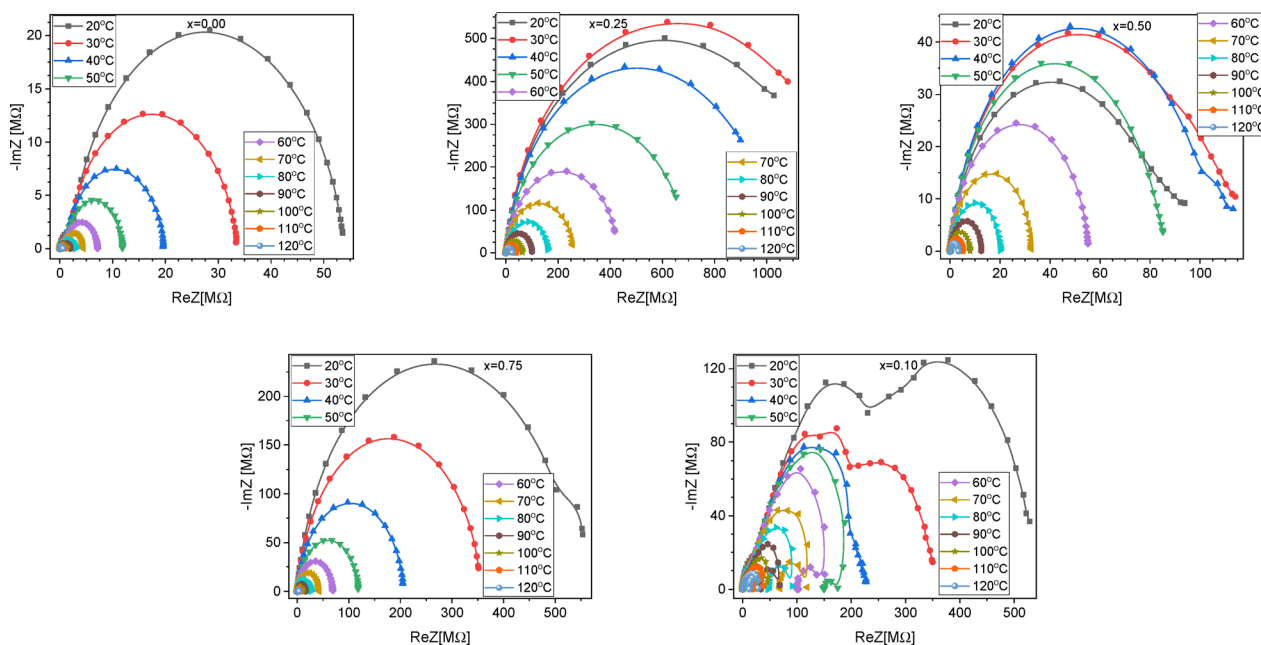


Fig. 15 Nyquist spectra of complex impedance ($-\text{Im}Z$ vs. $\text{Re}Z$) of the $\text{Sm} \rightarrow \text{MnNi}$ ($x \leq 0.10$) NSF.

in the Oh (B) regions, which reduces the overall resistance of the NSF network.^{14,18,19,22,23,37–39}

It is crucial to underscore that the Sm^{3+} -ion substitution ratio strongly influences the impedance response. The host NSF exhibits relatively large semicircular arcs at lower temperatures owing to the dominant grain boundary resistance. With a slight substitution ratio of 0.025, it is noteworthy that the arc diameter is further expanded, indicating increased grain boundary impedance resulting from defect states and localized potential barriers caused by the RE of Sm^{3+} ions.^{1,27,35,36}

In contrast to the above, the semicircular arc diameters are significantly reduced at moderate substitution levels in the range of $x = 0.050$ – 0.075 , indicating enhanced lattice distortion, improved hopping paths, and lower resistance. Furthermore, at the highest substitution level of $x = 0.10$, the arc diameter increases again, indicating that excessive Sm^{3+} -ion incorporation leads to defect clustering, lattice disorder, and carrier trapping centers that hinder charge transport. However, increasing the temperature moderately overcomes the corresponding potential barriers through thermal activation.^{18,19,22,37,38}

Therefore, it can be concluded that the single semicircular arcs confirm the dominance of grain boundaries in the conduction process and that the temperature- and composition-dependent impedance behavior shows that moderate Sm^{3+} -ion substitution increases electrical conduction, while excessive substitution impairs conductivity through defect-induced carrier localization.^{31,35,37,38}

3.12. Complex impedance: Nyquist fitting analysis

For a ferrite impedance spectrum, the three parallel branches can generally be discussed as distinct electrical relaxation/conduction processes, but their assignment to the grain, grain boundary, and an additional interfacial contribution should be based on the frequency ranges and the fitted Nyquist features, rather than simply assuming that R_1 , R_2 , and R_3 correspond to these regions, as summarized in Table 4. Therefore, the impedance spectra were fitted using the $R(\text{CR})(\text{QR})(\text{CR})$ equivalent circuit, consisting of a series resistance (R_s) and three parallel relaxation elements, namely $R_1 \parallel C_1$, $R_2 \parallel \text{CPE}$, and $R_3 \parallel C_3$. The constant-phase element was described by $Y_{\text{CPE}} = Y_0(j\omega)^n$, where Y_0 and n represent the CPE magnitude and the degree of deviation from ideal capacitive behavior, respectively.

The equivalent-circuit fitting results obtained using the $R(\text{CR})(\text{QR})(\text{CR})$ model reveal a pronounced dependence of the electrical response on both the substitution level and temperature. At 20 °C, the resistance parameters (R_s , R_1 , and R_2) vary by several orders of magnitude with x , indicating strong compositional modulation of the resistive and relaxation processes. Upon increasing the temperature to 120 °C, R_s generally decreases substantially, consistent with thermally activated charge transport in the ferrite system. The CPE exponent n also changes markedly with temperature, reaching values close to unity for several compositions, suggesting a progressively more ideal capacitive response and reduced non-Debye dispersion at elevated temperatures. The accompanying increase in Y_0 for several compositions further supports enhanced charge-carrier mobility and interfacial polarization at higher temperatures. However, the anomalously large/small fitted R and C values for some compositions should be treated cautiously, as they may reflect parameter correlation or limited fitting sensitivity rather than intrinsic physical quantities. Overall, the fitting results demonstrate that Sm substitution level and temperature strongly modify the resistive and relaxation components of the Mn–Ni ferrite impedance response, supporting thermally activated hopping and interfacial polarization mechanisms.

3.12.1. Non-Debye relaxation. The relaxation behavior was quantitatively evaluated using complex impedance/modulus analysis and the $R(\text{CR})(\text{QR})(\text{CR})$ equivalent-circuit model, incorporating constant-phase elements (CPEs) to account for non-ideal capacitive behavior. The observed depressed and broadened relaxation features, together with the fitted CPE exponents and temperature- and Sm-dependent relaxation parameters, indicate a distribution of relaxation times rather than a single ideal Debye relaxation. Thus, the non-Debye character is supported by the quantitative circuit analysis rather than being inferred solely from the shape of the impedance semicircles (Fig. 16).

3.13. Unified electrical mechanism

The electrical response is interpreted using the complementary information provided by conductivity, dielectric, impedance, and modulus analyses. Conductivity primarily reflects thermally activated localized charge transport and hopping, whereas dielectric behavior describes frequency- and temperature-dependent polarization. The impedance response provides evidence for electrically distinct grain and grain-boundary regions, while the electric

Table 4 Equivalent-circuit fitting parameters obtained from the impedance spectra of the Sm → MnNi NSFs with different substitution levels ($x \leq 0.100$) at (a) 20 and (b) 120 °C

Temp.	$T = 20\text{ °C}$					$T = 120\text{ °C}$				
Ratios, x	0.000	0.025	0.050	0.075	0.100	0.000	0.025	0.050	0.075	0.100
R_s (Ω)	2.2M	1.0G	4.1k	93.3k	404.6M	7.9k	1.6k	3.1m	0.47	
C_1 (F)	1.9p	1.0 μ	180.5n	138.4n	728.5n	>>	21.9p	>>	>>	
R_1 (Ω)	21m	1.5M	51 μ	34.8	23.0M	4.8	25.0M	824.1M	>>	
Y_0 (S s ^{n})	<<	8.8 μ	0.31	1.8n	2.1 μ	6.32p	101.2 μ	119.4p	51.8p	
n	<<	0.21	0.13	<<	0.38	0.97	6.7m	0.89	0.92	
R_2 (Ω)	0.73	7.5M	16.8u	8.0G	42.2M	622.5k	13.9	3.4M	4.6M	
C_3 (F)	32.3p	352.6n	26.7p	348.9 μ	713.3n	5.5 μ	702.7n	>>	<<	
R_3 (Ω)	48.0M	55.1M	80.5M	116.5M	100.5M	0.22T	540.2	>>	0.9	

Where “<<” → “too small” and “>>” → “too big”.

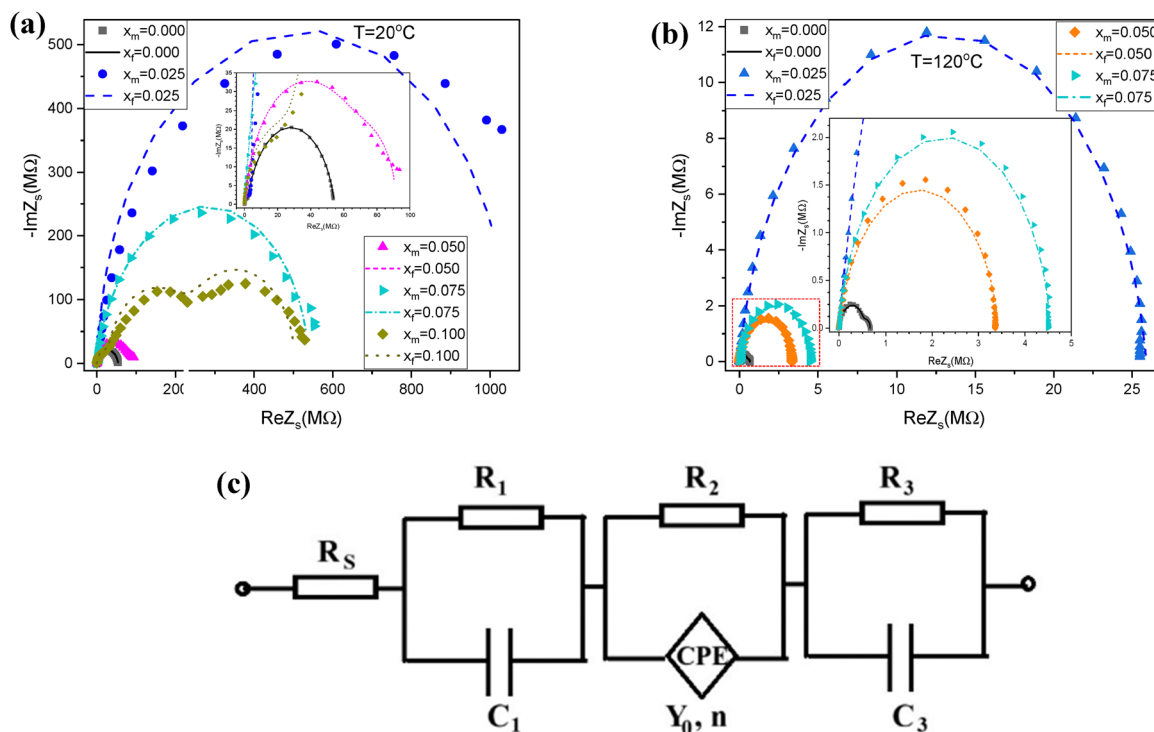


Fig. 16 Nyquist fitting analysis of the complex impedance of the Sm $\rightarrow$ MnNi NSFs at temperatures of (a) 20°C and (b) 120°C for some given substitution ratios (measured data and fitted lines). (c) Electrical equivalent circuit of the $R(\text{CR})(\text{QR})(\text{CR})$ fitting model.

modulus emphasizes localized relaxation processes and reduces the influence of electrode polarization. Accordingly, the low-frequency response is associated mainly with electrode/interfacial polarization, whereas grain and grain-boundary contributions become evident through the impedance response. The combined results indicate thermally activated hopping and interfacial polarization within an electrically heterogeneous system, while avoiding assignment of all observed features to a single microscopic mechanism.

The temperature- and frequency-dependent DC/AC conductivity, activation energy, frequency exponent, dielectric relaxation, and electric modulus responses indicate thermally activated localized charge transport and hopping between energetically accessible transition-metal sites. $\text{Fe}^{2+}/\text{Fe}^{3+}$ and $\text{Ni}^{2+}/\text{Ni}^{3+}$ hopping can therefore be considered physically plausible mixed-valence charge-transport pathways consistent with the observed electrical response. However, the present electrical and dielectric measurements do not directly determine the absolute concentrations or Sm-dependent evolution of the Fe and Ni oxidation states. Accordingly, these hopping processes are considered possible microscopic mechanisms rather than experimentally established oxidation-state changes.

The systematic variations in DC/AC conductivity, activation energy, frequency exponent, dielectric permittivity, dielectric loss, and electric modulus response indicate that Sm substitution modifies the localized charge-transport and relaxation behavior. These changes are consistent with modifications of local structural disorder, defect-related states, and cation environments, which may influence charge hopping and interfacial

polarization. However, the present electrical and dielectric measurements do not provide direct evidence for defect clustering, lattice distortion, or cation redistribution. Therefore, these effects are considered plausible contributing mechanisms rather than experimentally established structural phenomena.

4. Conclusion

The Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs were synthesized using the sol-gel approach. Their structure, morphology and chemical composition were analyzed *via* XRD, SEM, TEM, HR-TEM, EDX and elemental mapping. XRD analysis confirmed the cubic spinel ferrite formation. SEM, TEM and HR-TEM analyses showed the assembly of small spherical particles that form a porous structure. The present study demonstrates that the Sm $\rightarrow$ MnNi ($x \leq 0.10$) NSFs exert a significant influence on conduction dynamics and dielectric relaxation. All NSFs exhibit NTCR behavior, with thermally activated small-polaron hopping between ferrous and ferric ions at Oh (B) sites established as the dominant mechanism. Nyquist analysis of both impedance and modulus clearly confirms that conduction is primarily governed by grain boundary contributions and follows a non-Debye relaxation pattern. Moderate Sm^{3+} -ion substitution ($x = 0.050$ – 0.075) enhances conductivity and dielectric stability by optimizing lattice distortion and facilitating hopping pathways, whereas higher substitution ($x = 0.10$) introduces lattice disorder and defect clustering that hinder charge transport. The observed dual E_A regimes in DC conductivity further

confirm the coexistence of localized and long-range hopping transport. Overall, the obtained results demonstrate that controlled Sm^{3+} doping can effectively tune the electro-dielectric properties of MnNi-NSFs , making them promising candidates for high-frequency electronics and energy-storage applications.

Conflicts of interest

The authors declare no conflicts of interest regarding the publication of this article.

Data availability

The data produced and analyzed in this study can be made available from the corresponding author upon reasonable request.

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