

Structural, Optical and Dielectric Properties of $\text{Sm}_2\text{NiMnO}_6$ Double Perovskite Nanoparticles

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Abstract: $\text{Sm}_2\text{NiMnO}_6$ (SNMO) double perovskite was successfully synthesized via a sol-gel auto-combustion route followed by a post-combustion double sintering process to achieve phase purity and improved crystallinity. Rietveld refinement of PXRD data confirms the formation of a monoclinic structure with space group $\text{P2}_1/\text{n}$, indicating ordered arrangement of Ni^{2+} and Mn^{4+} cations at the B-site. Structural analysis reveals significant octahedral distortion, supported by a tolerance factor of ~ 0.923 and deviation of the Ni-O-Mn bond angle from 180° . Raman and FTIR studies further validate the formation of NiO_6 and MnO_6 octahedra and confirm lattice distortions at the local scale. FE-SEM analysis shows a nanostructured morphology consisting of agglomerated grains with noticeable porosity, which plays a crucial role in governing the dielectric behavior. Optical studies using UV-Vis diffuse reflectance spectroscopy indicate a direct allowed band gap of ~ 1.41 eV, suggesting potential applicability in optoelectronic and photovoltaic devices. The dielectric response of SNMO exhibits strong frequency dispersion in the range 5 kHz-1 MHz and temperature range 30-120 $^\circ\text{C}$. The dielectric constant decreases with increasing frequency, while it increases significantly with temperature, particularly at low frequencies, reaching values as high as $\sim 2.1 \times 10^4$ at 5 kHz and 120 $^\circ\text{C}$. This behavior is attributed to interfacial polarization arising from grain boundaries and pores, consistent with the Maxwell-Wagner model. The loss tangent also shows a similar trend, indicating increased conduction losses at elevated temperatures. At higher frequencies, the dielectric response stabilizes, reflecting intrinsic polarization mechanisms. The overall results demonstrate that the dielectric properties of SNMO are strongly influenced by its microstructure, with a transition from extrinsic to intrinsic behavior with increasing frequency.

Keywords: Double Perovskites, Maxwell-Wagner polarization, loss tangent.

1. Introduction

Double perovskite oxide materials represented as $AA'BB'O_6$ or $A_2BB'O_6$, have attracted considerable attention due to their rich structural flexibility and the emergence of diverse functional properties, including magnetism, dielectric behavior, magnetoresistance, and multiferroicity¹⁻⁵. These properties arise primarily from the ordered arrangement of two different transition metal cations at the B-site, which enables intricate electronic interactions and tunable physical characteristics with strong B–O–B' interactions. In particular, rare-earth-based double perovskites have become a focal point of research owing to the strong coupling between lattice, charge, and spin degrees of freedom, making them promising candidates for next-generation multifunctional devices in spintronics, optoelectronics, solar cells, electrocatalysis, sensing, photodetectors, and dielectric applications⁶⁻¹⁰.

Among various rare-earth double perovskites, R_2NiMnO_6 (R = rare-earth ion) have been extensively investigated due to their intriguing magnetic and dielectric properties¹¹⁻¹⁸. These materials belong to a special class of double perovskites in which a near-ideal $e_g^2 - O - e_g^0$ electronic configuration facilitates strong superexchange interactions between Ni^{2+} and Mn^{4+} ions. According to the Goodenough–Kanamori rules, this interaction is typically ferromagnetic (FM) in nature when an ordered arrangement of B-site cations is achieved.¹⁷ The magnetic ordering originates from virtual electron hopping between the half-filled orbital of Ni^{2+} and the empty orbital of Mn^{4+} . However, any deviation from ideal cation ordering, such as antisite disorder, introduces competing antiferromagnetic (AFM) interactions ($Ni^{2+}-O-Ni^{2+}$ and $Mn^{4+}-O-Mn^{4+}$), leading to magnetic frustration and complex magnetic states including spin-glass behavior^{19,20}.

Structurally, these systems can adopt different crystallographic symmetries depending on the degree of Ni/Mn ordering. A random distribution of B-site cations typically results in an orthorhombic phase, whereas an ordered arrangement stabilizes a monoclinic structure ($P2_1/n$)^{13,21}. The magnetic exchange interactions are highly sensitive to structural parameters such as Ni–O–Mn bond angles, bond lengths, and octahedral tilting. Furthermore, the magnetic properties are strongly influenced by the ionic radius of the rare-earth ion (R^{3+}). A decrease in ionic radii of R^{3+} enhances lattice distortion, weakens FM interactions, and reduces the Curie temperature²². In addition, increasing competition between nearest-neighbor FM and next-nearest-neighbor AFM interactions can drive a transition from FM ordering to an E*-type AFM structure with $\uparrow\uparrow\downarrow\downarrow$ spin alignment²³. The incorporation of rare-earth ions at the A-site further enriches the magnetic

behavior. Particularly, in $\text{Sm}_2\text{NiMnO}_6$, the presence of Sm^{3+} ions with a finite magnetic moment introduces additional magnetic interactions with the transition metal sublattice, resulting in phenomena such as spin reorientation, enhanced magnetic anisotropy, and increased coercivity at low temperatures^{24,25}. Structural distortions induced by the relatively smaller ionic radii of Sm^{3+} further modify the Ni–O–Mn interaction pathways, thereby influencing the overall magnetic response.

Beyond magnetism, R_2NiMnO_6 compounds are of significant interest due to their multifunctional nature, exhibiting properties such as magnetoresistance, magnetocapacitance, and semiconducting behavior. The dielectric response in these materials arises from multiple contributions, including intrinsic polarization, space charge effects, grain boundary contributions, and Maxwell–Wagner-type interfacial polarization^{25–29}. The presence of mixed valence states ($\text{Ni}^{3+}/\text{Mn}^{3+}$), oxygen vacancies, and charge carrier hopping further enhances dielectric dispersion, particularly at low frequencies. Notably, the large dielectric constant and relaxor-like behavior reported in systems such as $\text{La}_2\text{NiMnO}_6$ are closely associated with B-site cation ordering and charge distribution²⁸. The giant dielectric response is often attributed to charge ordering between Ni^{2+} and Mn^{4+} ions, while magnetodielectric effects near room temperature highlight the strong coupling between magnetic and dielectric properties^{30,31}.

Moreover, nanostructures offer an effective route to further tailor these properties. At reduced dimensions, increased surface-to-volume ratio, enhanced grain boundary effects, and size-induced strain significantly influence both magnetic and dielectric behavior. For instance, reduced particle size can broaden magnetic transitions, increase surface spin disorder, and modify dielectric relaxation dynamics. The sensitivity of the band gap and dielectric properties to synthesis conditions also indicates that structural distortions and electronic correlations play a crucial role in determining the overall functional response. Thus, the sol–gel auto-combustion method is particularly adopted due to its ability to ensure better chemical homogeneity, lower processing temperatures, and precise control over particle size and morphology. This method enables uniform mixing of precursor ions at the molecular level, which is essential for achieving high phase purity and optimal Ni/Mn ordering.

In this context, the present study focuses on the synthesis of $\text{Sm}_2\text{NiMnO}_6$ double perovskite nanostructures via a sol–gel auto-combustion route followed by a double-sintering protocol to enhance crystallinity and phase formation. The structural and dielectric properties of the synthesized material are systematically investigated. The study aims to provide deeper insights into

the correlation between structure and multifunctional properties in rare-earth-based double perovskite nanomaterials, thereby contributing to their potential applications in advanced electronic and spintronic devices.

2. Experimental Procedures

2.1 Synthesis of $\text{Sm}_2\text{NiMnO}_6$ Nanostructure

$\text{Sm}_2\text{NiMnO}_6$ double perovskite was synthesized using a sol-gel auto-combustion method employing a post-combustion double sintering protocol. Stoichiometric amounts of nickel acetate $[\text{Ni}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}]$, and manganese acetate $[\text{Mn}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}]$ were accurately weighed and dissolved in 40 mL double distilled water (DDW) under magnetic stirring at room temperature (Rt) to form clear precursor solutions. After complete dissolution of the acetate precursors, a stoichiometric amount of samarium nitrate $[\text{Sm}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}]$ was added. The resulting solution was continuously stirred to obtain a homogeneous metal-ion solution. Subsequently, citric acid was introduced as both a chelating agent and fuel, maintaining an appropriate molar ratio with respect to the total metal ions (3:1). The solution was then heated to $\sim 105^\circ\text{C}$ under constant stirring, along with the addition of 8 mL of ethylene glycol as a polymerizing agent. Heating was continued until a viscous gel formed due to solvent evaporation and complexation among the metal ions, citric acid, and ethylene glycol. Upon further heating, the gel underwent a self-sustained auto-combustion reaction, resulting in the formation of a voluminous and porous precursor powder due to rapid exothermic decomposition of the organic constituents and acetate groups. The obtained product was dried in a vacuum oven overnight and then ground thoroughly to improve homogeneity. The as-burnt powder was subsequently calcined at 650°C for 6 h and then at 1000°C for 7 h in air to achieve phase purity and crystallization of the double perovskite $\text{Sm}_2\text{NiMnO}_6$. After natural cooling to room temperature, the final product was reground to obtain a fine nanostructured powder.

2.2 Characterizations of Synthesized $\text{Sm}_2\text{NiMnO}_6$ Nanostructure

Powder X-ray diffraction (PXRD) pattern of the as-prepared sample was obtained using a Rigaku-Shimadzu smartlab instrument using Cu-K_α ($\lambda = 1.5406 \text{ \AA}$) radiation in the 2θ range from 10° to 90° with a scan speed of 1.5° per min. The collected diffraction data were analyzed by Rietveld refinement using the FullProf Suite. Refinement involved optimization of zero correction, scale factors, full width at half maximum (FWHM), asymmetry parameters, lattice parameters, thermal parameters, and fractional atomic coordinates. Background and peak shapes were simulated using linear

interpolation with refinable heights and a pseudo-Voigt function, respectively. Refinement parameters such as R_{wp} , R_p , R_{exp} and χ^2 were monitored throughout the iterative process. The refined crystal structures were visualized using VESTA software based on the generated CIF files. Raman spectroscopic analysis was conducted at room temperature using i-Raman Plus BWS465-532H spectrometer of Metrohm BWTEK with a 532 nm laser illumination. The surface morphology and elemental composition were examined using a field-emission scanning electron microscope (FE-SEM, Carl Zeiss, Germany) coupled with an energy-dispersive X-ray (EDX) spectrometer. Optical band gap energies were estimated from diffuse reflectance spectra (DRS) measured in the 200–2000 nm range using a JASCO V-770 UV-Vis-NIR spectrophotometer. The dielectric characteristics of the present ceramics were measured with a PSM1735 LCR-meter.

3. Result and Discussion

3.1 Structural, Vibrational and Morphological Study

The phase integrity and crystal structure of $\text{Sm}_2\text{NiMnO}_6$ (SNMO) were investigated using powder X-ray diffraction (PXRD) followed by Rietveld refinement, as shown in Fig. 1(a). The refinement was carried out using the Fullprof suite, and the close agreement between the observed (Y_{obs}) and calculated (Y_{calc}) diffraction profiles, along with acceptable goodness-of-fit parameter ($\chi^2 = 6.31$), confirms the high reliability of the refinement. The absence of any impurity peaks indicates the formation of a single-phase, highly pure compound. All diffraction peaks were successfully indexed to a monoclinic structure with space group $P2_1/n$, which is characteristic of ordered double perovskites. The presence of superlattice reflections further confirms the rock-salt ordering of Ni and Mn cations at the B-site. The refined lattice parameters are estimated as, $a = 5.35493 \text{ \AA}$, $b = 5.49843 \text{ \AA}$, and $c = 7.61056 \text{ \AA}$, indicating a deviation from ideal cubic symmetry. The sharp and intense diffraction peaks suggest good crystallinity of the synthesized compound. The structural stability and distortion were further analyzed using the Goldschmidt tolerance factor (t), according to the relation: $t = \frac{r_{\text{Sm}} + r_{\text{O}}}{\sqrt{2}(\frac{r_{\text{Ni}} + r_{\text{Mn}}}{2} + r_{\text{O}})}$, where r_{Sm} , r_{Ni} , r_{Mn} and r_{O} stands for the effective ionic radii of Sm, Ni, Mn and O ions, respectively. The tolerance factor (t) of SNMO was calculated using Shannon ionic radii, where $r_{\text{Sm}} = 1.24 \text{ \AA}$, $r_{\text{Ni}} = 0.69 \text{ \AA}$ (high spin), $r_{\text{Mn}} = 0.53 \text{ \AA}$, and $r_{\text{O}} = 1.40 \text{ \AA}$.²⁴ The obtained value ($t \approx 0.923$) is significantly less than unity, indicating a distorted perovskite structure and stabilization of the monoclinic phase.

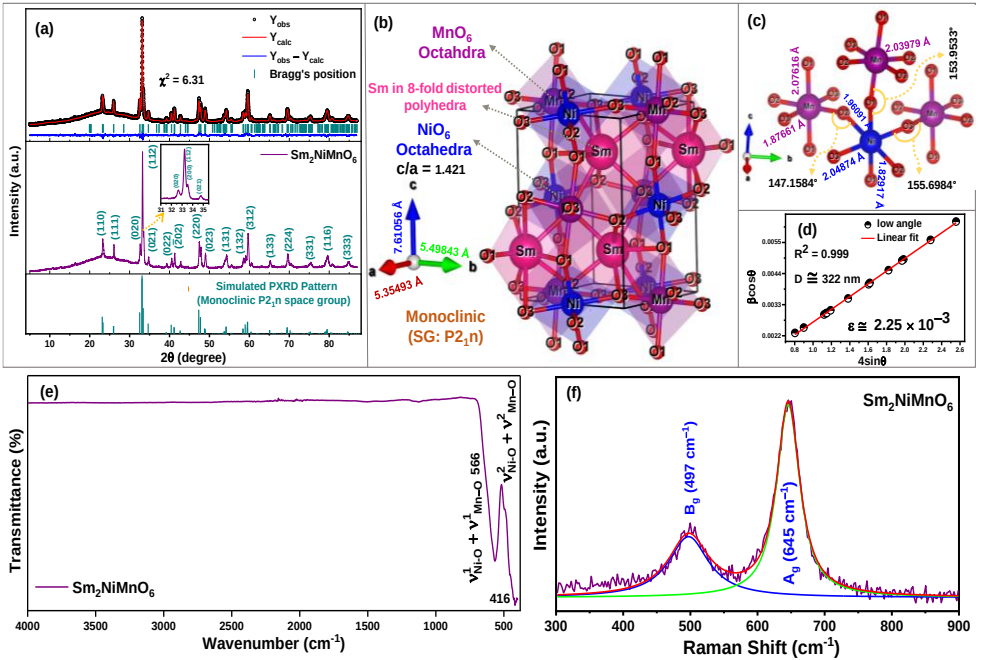


Fig. 1. (a) PXRD pattern with corresponding Rietveld refinement plot of $\text{Sm}_2\text{NiMnO}_6$ double perovskite (inset showing the enlarge view of most intense planes), (b) monoclinic unit cell structure, (c) crystallographic presentation of bond angles and bond lengths, and (d) Williamson-Hall plot used to estimate of crystallite size and microstrain, (e-f) Fourier Transform Infrared (FTIR) and Raman Spectroscopy at room temperature, respectively.

Table 1: Crystal structure parameters obtained from Rietveld refinement of PXRD pattern of $\text{Sm}_2\text{NiMnO}_6$.

Crystal structure: monoclinic, space group: $P2_1/n$ (#14), lattice parameter: $a = 5.35493 \text{ \AA}$, $b = 5.49843 \text{ \AA}$, $c = 7.610568 \text{ \AA}$, $\beta = 89.983^\circ$, unit cell volume: $V = 224.083081 \text{ \AA}^3$, structural tolerance factor (t) = 0.923, NiO_6 polyhedral volume = $9.7403 \text{ \AA}^3$, MnO_6 polyhedral volume = $10.5315 \text{ \AA}^3$, Ni effective coordination no. = 5.4016, Mn effective coordination no. = 5.4448, average bond angle (Θ) = 152.2733° , Octahedral tilt angle (Φ) = 13.863° . crystallite size (D) = 37 nm, micro strain= 1.14×10^{-3} , Conventional R-factors: R_{wp} (%) = 15.0, R_{exp} (%) = 5.98, R_p (%) = 25.1, $\chi^2 = 6.31$				
Atom	Wyckoff	x	y	z
Sm	4e	0.9872	0.0542	0.2483
Ni	2c	0.5000	0.0000	0.5000
Mn	2d	0.5000	0.0000	0.0000
O1	4e	0.0806	0.4861	0.2333
O2	4e	0.7241	0.2849	0.0676
O3	4e	0.7216	0.2993	0.4626

Ni–O bond length (Å)	Mn–O bond length (Å)	Bond angles (deg.)
Ni–O1 ($\times 2$) = 1.82917	Mn–O1 ($\times 2$) = 2.07616	$\angle \text{Ni–O1–Mn} = 153.9633^\circ$
Ni–O2 ($\times 2$) = 1.96091	Mn–O2 ($\times 2$) = 2.03979	$\angle \text{Ni–O2–Mn} = 147.1584^\circ$
Ni–O3 ($\times 2$) = 2.4874	Mn–O3 ($\times 2$) = 1.87661	$\angle \text{Ni–O3–Mn} = 155.6984^\circ$
$\langle \text{Ni–O} \rangle = 1.9463$	$\langle \text{Mn–O} \rangle = 1.9975$	$\langle \angle \text{Ni–O–Mn} \rangle =$ 152.2733°

The refined crystal structure, depicted in Fig. 1(b), reveals that Sm^{3+} ions occupy the A-site (Wyckoff position 4e), while Ni and Mn ions are located at the 2c and 2d sites, respectively, with oxygen atoms occupying the 4e positions. Both Ni and Mn ions are coordinated by six oxygen atoms, forming NiO_6 and MnO_6 octahedra that are alternately arranged in a rock-salt-type configuration. These octahedra are corner-shared via oxygen atoms, forming a three-dimensional framework. The Sm^{3+} ions are surrounded by eight oxygen atoms, resulting in a distorted polyhedral environment due to the tilting of adjacent $\text{MnO}_6/\text{NiO}_6$ octahedra. The detailed bond geometry as shown Fig. 1(c), indicates that the average Ni–O and Mn–O bond lengths are ~ 1.946 Å and ~ 1.997 Å, respectively. It indicates the expected oxidation states of Ni^{2+} and Mn^{4+} . The Ni–O–Mn bond angles deviate significantly from the ideal 180° , with an average value of $\sim 152.27^\circ$, indicating pronounced octahedral tilting. The corresponding octahedral tilt angle (Φ) is 13.86° , calculated using $\Phi = (180 - \Theta)/2$, confirming the presence of structural distortion arising from the size mismatch and ordering of B-site cations. Furthermore, the crystallite size and lattice strain were evaluated using both the Scherrer and Williamson–Hall (W–H) methods. The W–H plot was constructed by plotting $\beta \cos \theta$ as a function of $4 \sin \theta$ and fitted linearly to obtain lattice strain (ϵ) of $\sim 2.26 \times 10^{-3}$, indicating the presence of a small but finite lattice distortion. The crystallite size obtained from the W–H intercept was ~ 322 nm, suggesting relatively large coherent scattering domains. In contrast, the crystallite size calculated using the Scherrer formula was found to be ~ 53 nm, which is more representative of the nanoscale domain size. The discrepancy between these two values arises because the Scherrer method accounts only for size-induced broadening, whereas the W–H method separates both size and strain contributions. The comparatively larger crystallite size obtained from the W–H analysis suggests that peak broadening is significantly influenced by strain and possible instrumental effects, leading to an overestimation of crystallite size.

Moreover, the FTIR spectrum of $\text{Sm}_2\text{NiMnO}_6$ as depicted in Fig. 1e exhibits characteristic features typical of double perovskite oxides and is consistent with previously reported R_2NiMnO_6 double perovskites (Table 2). Two prominent absorption bands are observed in the low-wavenumber

region, confirming the formation of the perovskite structure. The higher-frequency band around $\sim 566 \text{ cm}^{-1}$ is attributed to the combined anharmonic stretching vibration of the Ni–O and Ni–O within the octahedral structural unit, while the lower-frequency band near $\sim 416 \text{ cm}^{-1}$ corresponds to the octahedral deformation bending mode of Ni–O/Mn–O bonds^{32–34}. The presence and positions of these bands indicate the formation of well-defined NiO₆ and MnO₆ octahedra. The slight variation in vibrational frequencies compared to other R₂NiMnO₆ compounds can be associated with differences in octahedral environment due to different rare-earth elements.

Table 2. comparison of the observed vibrational frequencies with the reported studies for the different rare-earth element based R₂NiMnO₆ double perovskites

Compound	Z of rare-earth element	Ionic radii of rare-earth element (Å)	ν^1 (cm ⁻¹)	ν^2 (cm ⁻¹)	Reference
La ₂ NiMnO ₆	57	1.16	430	600	Ahmad et al. ³⁵
Y ₂ NiMnO ₆	39	1.019	464	586	Lei et al. ³⁶
Pr ₂ NiMnO ₆	59	1.126	440	590	Elhamel et al. ³⁷
Nd ₂ NiMnO ₆			430	565	Hossain et al. ³⁸
Eu ₂ NiMnO ₆	63	1.066	–	602	Elhamel et al. ³⁹
Gd ₂ NiMnO ₆	64	1.053	460	595	Elhamel et al. ³⁷
Ho ₂ NiMnO ₆	67	1.015	445	560	Bhat et al. ⁴⁰
Er ₂ NiMnO ₆	68	1.004	470	602	Elhamel et al. ³⁷
Sm ₂ NiMnO ₆	62	1.079	–	575	Mukherjee et al. ³²
			416	566	In this work

Meanwhile, Raman spectroscopy was further employed to investigate the local structure and phonon dynamics of Sm₂NiMnO₆ at room temperature as represented in Fig. 1f. Based on the monoclinic P2₁/n (*C*_{2h}⁵ point group) symmetry, there is 24 zone-center Raman active modes and represented as: $\Gamma_{\text{Raman}} = 6T(3A_g + 3B_g) + 6L(3A_g + 3B_g) + 2\nu_1(A_g + B_g) + 4\nu_2(2A_g + 2B_g) + 6\nu_5(3A_g + 3B_g)$.^{32,41} Here, L and T denote the vibrational and translational phonon modes, respectively. While ν_1 , ν_2 , and ν_5 correspond to the symmetric stretching, antisymmetric stretching, and symmetric bending vibrations of the NiO₆ (weak) and/or MnO₆ (strong) octahedra. In Fig. 1f, the Raman spectrum shows two dominant modes at 645 cm^{-1} (*A*_g) and 497 cm^{-1} (*B*_g), attributed to symmetric stretching (breathing) and mixed anti-stretching/bending vibrations of the octahedra, respectively. The spectrum is well fitted using Lorentzian functions. The observed peak broadening indicates phonon confinement and participation of non-zone-center phonons. This indicates the characteristic of nanocrystalline features and suggests the presence of structural disorder and lattice distortion⁴².

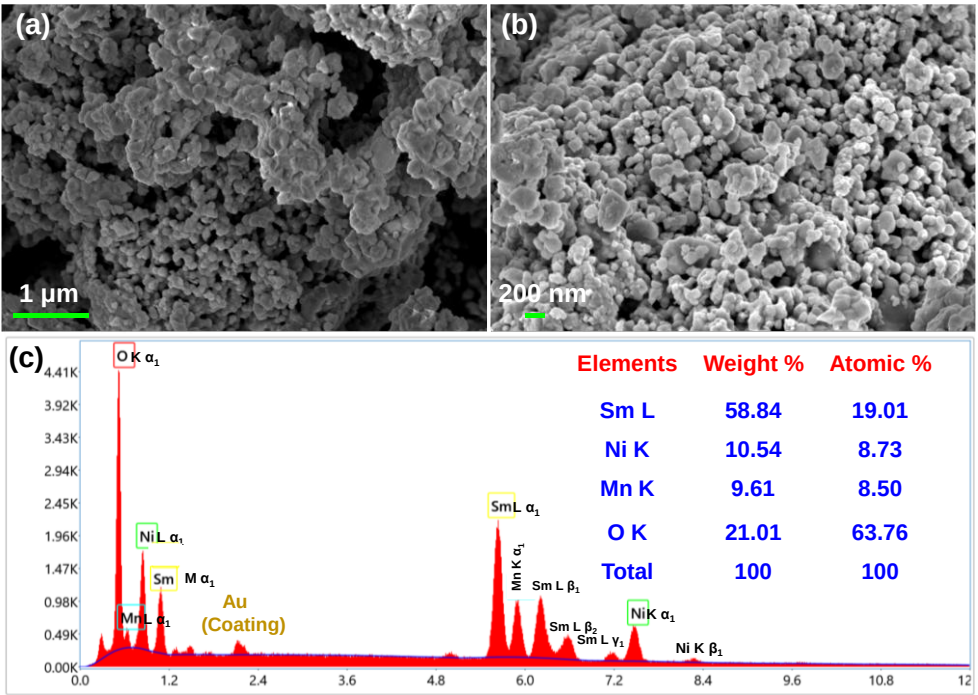


Fig. 2(a–b) SEM images of $\text{Sm}_2\text{NiMnO}_6$ at 43.3 k $\times$, and 53.6 k $\times$ magnifications, respectively; and (c) Energy-Dispersive X-ray Spectroscopy (EDXS) with element’s Weight% and Atomic%.

The SEM images as shown in Fig. 2a–b, reveal that $\text{Sm}_2\text{NiMnO}_6$ exhibits a highly agglomerated morphology composed of nearly spherical nanograins. At higher magnification, the particles are densely packed with irregular boundaries, indicating nanoscale crystallites with significant grain clustering, which is typical for sol–gel derived materials. The EDX spectrum as presented in Fig. 2c, confirms the presence of Sm, Ni, Mn, and O without any detectable impurity phases, indicating good phase purity. The elemental composition (Sm: 58.84 wt%, Ni: 10.54 wt%, Mn: 9.61 wt%, O: 21.01 wt%) is in reasonable agreement with the expected stoichiometry. The uniform distribution of elements suggests successful formation of the desired $\text{Sm}_2\text{NiMnO}_6$ double perovskite.

3.2 Optical Properties: UV-Vis diffuse reflectance spectroscopy (DRS)

The optical properties of $\text{Sm}_2\text{NiMnO}_6$ (SNMO) were investigated using UV–Vis diffuse reflectance spectroscopy (DRS) at room temperature, as shown in Fig. 3(a). The spectrum exhibits broad absorption extending from the visible to the near-infrared region, which arises from electronic transitions between the valence band and conduction band states of SNMO. These transitions are primarily governed by strong hybridization between Ni/Mn–3d and O–2p

orbitals, along with minor contributions from Sm–4f states, reflecting the mixed covalent–ionic nature of bonding in the system^{43,44}.

The diffuse reflectance data were analyzed using the Kubelka–Munk Function, $F(R) = \frac{(1-R)^2}{2R}$, where R is the diffuse reflectance and the

corresponding variation of F(R) is presented in Fig. 3(b). The absorption edge confirms the semiconducting nature of the material in the visible region. The absorption edge cut-off wavelength, obtained from extrapolation to the wavelength axis in the plot of $[F(R)hv]$ with wavelength, is approximately 942 nm. The optical band gap energy (E_g) was initially estimated from the absorption edge using the relation $E_g = hc/\lambda$. Upon substitution of fundamental constants and conversion into electron volts, reduces to $E_g = 1240/\lambda$. This yields a band gap value of approximately 1.31 eV. For a more accurate determination of the optical band gap, Tauc analysis based on the Kubelka–Munk (K–M) function was employed, considering the presence of band-tail states near the valence and conduction band edges. The analysis

follows the relation: $[F(R)hv]^{\frac{1}{n}} = C_1(E_g - hv)$, where hv is the photon energy, C_1 is a proportionality constant and n characterizes the nature of the optical transition. The exponent n takes values of 3, 2, 3/2, and 1/2 for indirect forbidden, indirect allowed, direct forbidden, and direct allowed transitions, respectively.^{45,46} Among these, the plot with $n = 1/2$ exhibits the best linear fit,

indicating that the optical transition in the present system is predominantly direct allowed in nature. The extracted band gap is found to be 1.41 eV as shown in Fig. 3(c), which is in close agreement with reported theoretical calculations (1.38 eV) based on spin-up channel DOS analysis.⁴⁴ Interestingly, this band gap is very close to the Shockley–Queisser limit (~1.34–1.39 eV) for maximum efficiency in single-junction solar cells, suggesting that SNMO can be a promising candidate for photovoltaic applications.⁴⁷ The relatively narrow band gap is attributed to strong (Ni/Mn)-3d and O-2p hybridization, which defines the valence band maximum, while the conduction band is mainly composed of transition metal d-states. Further insight into the degree of structural disorder was obtained from Urbach

energy (E_u) analysis using the relation $\alpha = \alpha_0 \exp(\frac{hv}{E_u})$, α stand for

absorption co-efficient, hv is the photon energy, α_0 is a constant and shown in Fig. 3(d). The Urbach energy reflects the extent of band tailing due to localized states arising from defects such as oxygen vacancies and lattice disorder. The moderate Urbach energy (557 meV) value indicates the presence of some degree of structural disorder, which contributes to band tailing and enhanced sub-bandgap absorption. Overall, the optical response of SNMO is governed by its electronic structure, particularly the strong

hybridization between Ni/Mn–O orbitals, along with defect-induced localized states. The combination of a narrow direct band gap, favorable electronic structure, and moderate disorder makes SNMO a potential material for optoelectronic and photovoltaic applications.

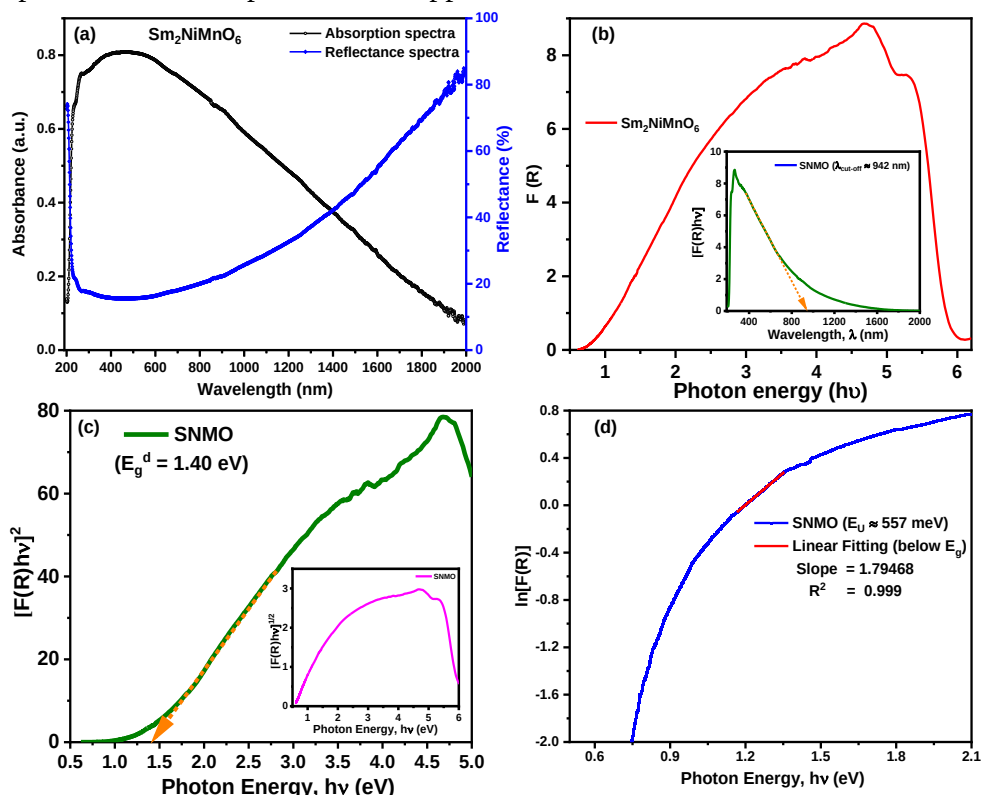


Fig. 3 (a) Samples' UV-Vis DRS spectra, (b) variation of Kubelka–Munk, $F(R)$ function with photon energy and inset showing the plot of $[F(R)hv]$ with wavelength for the estimation absorption edge cut-off wavelength, (c) Plot of $[F(R)hv]^2$ vs $h\nu$ to estimate energy band gap and inset showing the plot of $[F(R)hv]^{1/2}$ vs $h\nu$, (d) $\ln[F(R)]$ vs $h\nu$ plot to calculate Urbach energy.

3.3 Frequency-Dependent Dielectric Response with Temperature Variation

The frequency-dependent dielectric parameters from 5 kHz to 1 MHz with temperature variation in a range of temperature 30 °C–120 °C with a step of 30 °C, of double-perovskite compositions SNMO were examined through standard relations. Frequency dependent dielectric constant (ϵ) and tangent loss ($\tan\delta$) observed with temperature variation in the temperature range 30 °C–120 °C are shown in Fig. 4. A strong frequency dispersion is observed in both ϵ and $\tan\delta$, which is characteristic of heterogeneous dielectric systems. At 30 °C, the dielectric constant decreases from 484 at 5

kHz to 224 at 1 MHz, indicating a gradual suppression of polarization mechanisms with increasing frequency. At elevated temperature (120 °C), a significantly enhanced dielectric response is observed at low frequency, where ϵ decreases sharply from 21479 at 5 kHz to 267 at 1 MHz similar to previous reports^{48,49}. This pronounced reduction with frequency suggests that the dominant contribution at low frequency arises from interfacial polarization, which becomes ineffective at higher frequencies⁵⁰. The observed dielectric dispersion can be explained on the basis of Maxwell–Wagner polarization, which originates from the accumulation of charge carriers at interfaces such as grain boundaries and pores⁴³. This is consistent with the SEM analysis, which reveals a nanograined and porous microstructure, providing a large density of interfaces for space charge accumulation. The temperature dependence further supports this mechanism. The substantial increase in ϵ at low frequencies with temperature is attributed to thermally activated charge carriers, which enhance interfacial polarization and space charge effects. In contrast, at higher frequencies (≥ 100 kHz), ϵ shows relatively weak temperature dependence, approaching intrinsic dielectric behavior dominated by ionic and electronic polarization.

The loss tangent ($\tan\delta$) exhibits a similar frequency-dependent trend. At 30 °C, $\tan\delta$ decreases from 0.61971 at 5 kHz to 0.07457 at 1 MHz, indicating relatively low dielectric loss at higher frequencies. However, at 120 °C, $\tan\delta$ increases significantly at low frequency (6.2924 at 5 kHz) and decreases to 0.17503 at 1 MHz. The high loss at low frequency and elevated temperature is indicative of increased conduction losses arising from leakage current and hopping charge transport between transition metal ions⁵¹.

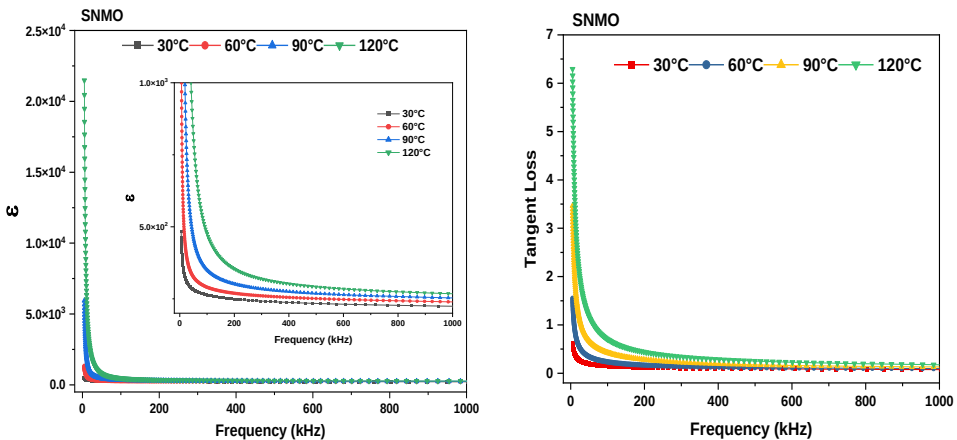


Fig. 4: Temperature-dependent a) dielectric constant and b) tangent loss of SNMO

Overall, the dielectric behavior of SMNO demonstrates a transition from extrinsic, interface-dominated polarization at low frequencies to intrinsic

dielectric response at high frequencies. The large dielectric constant observed at low frequency and high temperature is therefore not purely intrinsic but strongly influenced by microstructural effects, particularly grain boundaries and porosity.

4. Conclusions

In summary, $\text{Sm}_2\text{NiMnO}_6$ double perovskite was successfully synthesized using a sol-gel auto-combustion method followed by optimized double sintering, leading to the formation of a single-phase monoclinic structure with space group $\text{P}2_1/\text{n}$. Structural analysis confirms ordered B-site cation arrangement along with significant octahedral distortion arising from size mismatch and tilting of NiO_6 and MnO_6 octahedra. Vibrational studies through Raman and FTIR spectroscopy further corroborate the structural framework and reveal lattice disorder associated with nanocrystallinity. Morphological analysis demonstrates a nanograined and porous microstructure, which plays a decisive role in determining the dielectric response. Optical investigations reveal a narrow direct band gap (~ 1.41 eV), indicating potential applicability in optoelectronic and photovoltaic devices. The dielectric study reveals strong frequency and temperature dependence, characterized by a decrease in dielectric constant with increasing frequency and a significant enhancement at elevated temperatures, particularly in the low-frequency region. The large dielectric constant observed at low frequencies is primarily attributed to interfacial polarization and space charge effects associated with grain boundaries and pores, consistent with the Maxwell-Wagner mechanism. The accompanying increase in dielectric loss at high temperature further indicates the contribution of thermally activated charge carriers and leakage conduction. At higher frequencies, the dielectric response becomes relatively stable, reflecting intrinsic polarization mechanisms. Thus, the dielectric behavior of SNMO demonstrates a clear transition from extrinsic, microstructure-driven effects at low frequencies to intrinsic dielectric response at high frequencies. These findings highlight the critical role of microstructure in tuning the functional properties of double perovskite oxides and establish SNMO as a promising material for dielectric and optoelectronic applications.

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