Studies of structural, dielectric and electrical characteristics of complex perovskite: Sr(Ni$_{1/3}$Mn$_{1/3}$W$_{1/3}$)O$_3$

D Khatua$^1$, S K Dehury$^2$, S K Parida$^3$, R N P Choudhary$^3$ and P G R Achary$^{1*}$

$^1$Department of Chemistry, Siksha ‘O’ Anusandhan Deemed to be University, Khandagiri Square, Bhubaneswar 751030, India
$^2$P.G. Department of Chemistry, Vikram Deb (Autonomous) College, Jeypore 764001, India
$^3$Department of Physics, Siksha ‘O’ Anusandhan Deemed to be University, Khandagiri Square, Bhubaneswar 751030, India

Received: 22 June 2019 / Accepted: 24 January 2020 / Published online: 3 July 2020

Abstract: A lead-free complex perovskite, strontium nickel tungsten manganite Sr(Ni$_{1/3}$Mn$_{1/3}$W$_{1/3}$)O$_3$(SNMWO), was synthesized following a cost-effective standard ceramic processing technology. The structure of the compound was found to be cubic. A detailed study of scanning electron micrograph and the energy-dispersive spectra of SNMWO provided the quality and composition of the material. The average crystallite size ($D_{hlkl}$) of SNMWO was found to be about 22 nm, which was calculated using Scherrer relation. The analysis of the data of dielectric, impedance and electrical conduction for a wide range of frequency and temperature of SNMWO provides interesting characteristics for useful applications. The dielectric relaxation and electrical properties are related to the grains, grain boundaries and electrode characteristics of SNMOW. Such characteristics of the material help us to understand the correlation between conduction mechanism and microstructure of the material for electronic applications. The important electrical properties were evaluated from the simulation of the Nyquist plots with the help of an equivalent electrical circuit. The transport properties along with the ac-conductivity, modulus (electrical) of Sr (Ni$_{1/3}$Mn$_{1/3}$W$_{1/3}$)O$_3$, were also presented in the paper.

Keywords: Perovskite; Multiferroic material; Electrical properties; Nyquist plots; Dielectric relaxation

PACS Nos.: 61.05.C--; 77.22.Gm

1. Introduction

The compound having ABX$_3$ (A = mono or divalent, B = tri to hexavalent ions)-type formula with greater size difference between the cations (A and B) and the anion(X) has a perovskite structure. These perovskites display greater flexibility in their structure and often referred as ‘inorganic chameleon’. In most of the cases, we find the anion X as an oxide, but it could be S$^{2-}$, Cl$^{-}$, Br$^{-}$, I$^{-}$ or N$_3^-$ as well. The A-site cations such as Pb$^{2+}$, Sr$^{2+}$, Ca$^{2+}$ occupy the corner sites and the transitions metal cations, like Ni$^{2+}$, Mn$^{4+}$, W$^{6+}$, etc., which have the incomplete d-electrons occupy the B-sites [1]. The introduction of the more ions at the A- and/or B-sites distorts the crystal structure of the perovskite. Also, a single perovskite can be changed to a double or complex perovskite structure with a general formula A$_1$A$_2$B$_1$B$_2$O$_6$ [2]. Such type of distortions in the crystal structure can have a dramatic effect on the electrical, optical, elastic or other specific characteristics of the material [3]. Among the different perovskites, the manganite perovskites are potential candidates for many devices including thermo-electric devices [4].

Perovskite manganites having working formula AMnO$_3$, where A = Ca$^{2+}$, Ba$^{2+}$ or Sr$^{2+}$ or alkaline earth metal ion, are considered as suitable candidates to exhibit better electrical and magnetic characteristics [5]. These materials have an added advantage, where we can alter the properties of materials by changing the magnetic or electric fields; this gives an added mode of control to tune technological applications in the area of spintronic devices, magnetic sensors, catalysts, magnetoresistive, magnetic recording devices, transducers and magnetic refrigeration.

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s12648-020-01789-1) contains supplementary material, which is available to authorized users.

*Corresponding author, E-mail: pgrachary@gmail.com

© 2020 IACS
systems [6–8]. The chemical and physical properties of perovskite oxides vary with the composition from dielectric to superconducting and find different applications [5]. Perovskite manganites when doped can exhibit better ferromagnetism around room temperature, such manganites attracted researchers in the recent past for the applications in devices like magnetic field sensors, high-density memory applications and infrared detectors. For example, La_{1−x}Sr_{x}MnO_{3} manganite films have given due attention for their remarkable property toward the spin-dependent tunneling and colossal magnetoresistance (CMR) effect [9]. If these perovskites could be integrated on silicon, they can find applications in the semiconductor industries. If one compares with other electroactive perovskite oxides, these manganite perovskites are more sensitive because of the lattice distortion.

Recently, owing to some attractive properties of the perovskites, serious attempts have been made to alter the perovskite structure by suitable substitution at the A/B-sites or fabricating solid solution/composites [10]. With such type of modification in the structure of the perovskites, researchers have always aimed to tune the characteristics to find applications in smart electronic devices, sensors for clinical devices, etc. In the love for the environment and to make each process green, we always look for the lead-free devices better than the PZTs.

SrMnO_{3} is an interesting lead-free manganite with perovskite structure, which possesses both cubic and hexagonal perovskite polymorph. SrMnO_{3} arranges in cubic form due to the octahedral sharing of MnO_{6} at a corner, whereas in hexagonal form is build due to the sharing of con facial dioctahedral sharing of Mn_{2}O_{6} in the direction of c-axis. Both cubic and hexagonal SrMnO_{3} forms are insulators having band gaps of 0.3 and 1.6 eV, respectively. The stability of the hexagonal SrMnO_{3} could be maintained up to 1035 °C and above this temperature, it changes to cubic SrMnO_{3}. The enthalpy of this transition is about 6 ± 2 kJ mol^{-1} [11]. However, the cubic SrMnO_{3} can be quenched and maintained as a metastable compound at low temperatures. This cubic structure is known to be a G-type antiferromagnetic material having around 260 meV more energy than the hexagonal polymorph. The stability of both of these forms can be understood by the ionic interaction of Sr^{2+} and the covalent character of the Mn–O bond. The stability of hexagonal SrMnO_{3} is explained by two factors: (1) the displacement of Mn atom along the c-axis from the center and (2) lower charge of O atoms at face sharing triangle than the corner-sharing O-atom. The change in negative charge of O atoms balances the repulsion of Mn–Mn in the Mn_{2}O_{6} dimer justifies the short Mn–Mn bond distance in the hexagonal SrMnO_{3}. Some of the manganites with perovskite structure are modified as: SrCoO_{3−δ} [SrCo_{1−x}Mo_{x}O_{3−δ}] [12]. Sr(Mn_{1−x}Mo_{x})O_{3} [13]. The effect of doping in SrMnO_{3} perovskite nanofibers was studied by George et al. The authors doped Ba/Ca on Sr and Co/Fe/Ni on Mn site of the perovskite and studied the electrical impedance and specific conductance of the doped perovskite. The 20 mol % Ba doped in SrMnO_{3} has high capacitive properties, and it is reported that the modified manganite has better supercapacitor application [14]. The dielectric, electrical study on lead-based perovskite Pb(Ni_{1/3}Mo_{1/3})O_{3} reported in our previous communication [15].

The doping or substitution of Ni and W in SrMnO_{3} at the B-site is expected to display interesting properties in the structure as well as in the dielectric properties. So, such type of materials find applications in different shape memory/or spintronic devices [16]. We have already reported the synthesis and characterization of some complex perovskites: Bi_{0.5}Pr_{0.5}Fe_{0.5}Co_{0.5}O_{3} [17], Bi_{0.5}Sr_{0.5}Fe_{0.5}Co_{0.5}O_{3} [18], BiBaFeCoO_{3} [19], BiHoCoTiO_{6} [20], BiHoZnZrO_{6} [21] and BiHoCuTiO_{6} [21].

The present investigation aims at the dielectric, electrical and structural study of strontium manganite modified by nickel and tungsten at the B-Site [Sr(Ni_{1/3}Mn_{1/3}W_{1/3})O_{3}], with perovskite structure, which is expected to show a ferroelectric transition.

2. Experimental

The complex strontium manganite (SNMWO) was synthesized by adopting a cost-effective solid-state reaction mechanism by taking the stoichiometric proportions of analytical grades of SrCO_{3}(> 96%, Sigma-Aldrich), NiO (99.99%, Merck India), MnO_{2} (70%, Merck India) and WO_{3} (99.99%, LobaChemie). The above metal oxides and carbonates were taken in an agate mortar and pestle and ground manually for 2.5 h followed by wet grinding with the methanol for an hour to get a homogeneous mixture of the above compounds. Then, the above mixture was taken in a cleaned cylindrical alumina crucible and placed in the temperature-programmed furnace at different temperatures starting from 600 to 1100 °C. The formation of SNMWO is confirmed by analyzing the X-ray diffraction data collected on the powder sample using a powder diffractometer (model D8 Advance of M/s Bruker Co) with wavelength 1.5405Å and 2θ ranges from 20° to 80°. The pellets of SNMWO were prepared of 10 mm diameter and thickness around 2 mm and placed in the high-temperature furnace again on a clean alumina boat for sintering at 1130 °C for 10 h. Then, the SNMWO pellets were cooled to 25 °C (1 day) to avoid any type of thermal shock. The sintered pellets were taken from the oven and verified its texture and breakage to ensure proper sintering of the pellet. One such properly sintered pellet was gold coated.
and sent for the morphology study using an SEM apparatus having model JEOL–JSM-6400 at 20 kV. The second pellet was made conducting both sides by applying uniform Ag-paint (high conducting) and put it in a hot air oven at 410 K (2 h) to check the removal of moisture or impurities (volatile). The continuity of the electrode on the two faces of the pellet was also verified before the electrical and dielectric analysis of SNMWO perovskite. The electrical and dielectric analysis of SNMWO sample was carried out using the LCR [impedance analyzer or phase-sensitive meter (PSM)]. Various electrical parameters were measured in the temperature range of 25 to 500 °C and frequency range of 100 Hz to 1 MHz.

3. Results and discussion

3.1. Crystal structure and morphology

Figure 1(b) represents the powder X-ray diffraction pattern of SNMWO with several distinct reflections. The sharp peaks of the XRD pattern show that the prepared SNMWO is single-phase polycrystalline material. The crystallite size ($D_{hkl}$) of SNMWO is determined from the XRD reflection pattern [Fig. 1(a)] by using Debye–Scherrer relation;  

$$D_{hkl} = \frac{K \lambda}{\beta_{0.5} \cos \theta},$$

where $K$ = constant $= 0.89$, $\lambda = 1.5418$ Å and $\beta_{0.5}$ is full width at half maximum (FWHM) [22]. The average crystallite size of SNMWO was found to be 28 nm which was determined from the broadening of some of the prominent peaks by avoiding the stain effect. The refinement of the crystal structure of the material was done by GSAS-II software code using the XRD diffraction data. $\text{Sr}_2\text{NiWO}_6$ with cubic structure (space group Fm-3 m) is taken as a reference model/pattern (source: crystallography open database (COD)). The initial values of cell parameter were: $a = 7.85897$ Å, cell vol = 485.397 Å$^3$ and density = 7.031 (g/cm$^3$). The Pseudo-Voigt function describes the Fourier series with the refinable coefficients. The Pseudo-Voigt function is the linear combination of Lorentzian and Gaussian functions which can be used to resolve strain and size contributions to the peak broadening [23]. It can be clearly observed in Fig. 1(b) that the diffraction peaks (reflection) and the intensities of the sample are perfectly matching with that of the model. The diffraction peak showing with the ‘+’ symbol represents the XRD data of SNMWO. The continuous line represents the calculated (above) and the difference (below) curves, and the vertical bars represent the Bragg’s reflections of the crystalline phase. The Bragg’s reflections indicate that the SNMWO sample is a single phase with a cubic structure. The Rietveld refined pattern confirms the cubic structure (space group Fm-3 m) of the SNMWO sample, and the refined structural parameters are found to be: $a = 7.9182$ Å, cell volume = 496.456 Å$^3$ and density 6.874 (g/cm 3) with $R_w$ less than 10%.

![Fig. 1](a) SEM micrograph of SNMW, (b) Rietveld refinement of SNMW; (c) The elemental composition of SNMWO and (d) EDS layered mapping of the distribution of the elements
The SEM micrograph of SNMWO was represented in Fig. 1(b). The micrograph of SNMWO shows elongated grains with a size variation of 0.6 μm to 2 μm. Energy-dispersive X-ray spectroscopy (EDX or EDS) in combination with SEM on the small area of the sample SNMWO provided the elemental composition of the sample. The energy-dispersive spectra (EDX) of the SNMWO is given in Fig. 1(c) which confirms the presence of all the elements in the required stoichiometric proportion (weight percentages) in SNMWO sample. The colored EDS layer mapping is shown as an inset in Fig. 1(d), which displays the uniform distribution of elements in the sample.

3.2. Dielectric properties

At any particular frequency, the ratio of the impedance of the real and imaginary component of any material, which is behaving as a capacitor, is known as dielectric loss or loss tangent (tan δ). The high value of tan δ brings high dielectric absorption. For good dielectric material, the capacitance changes slowly with the change in frequency. Hence, the value of tan δ can be calculated from the rate of decline in the capacitance. The relative dielectric constant \( \varepsilon_r \) is usually expressed as the condenser capacity of the sample divided by the capacity of the vacuum as the dielectric medium. The \( \varepsilon_r \) generally deduced from the formalism: \( \varepsilon_r = C_p/C_0 \), where \( C_p \) means parallel \( C \) which can be obtained from the impedance analyzer data and \( C_0 = (\varepsilon_o A/d) \) with \( A \) as area and \( d \) being the thickness of the sintered pellet.

The variation of \( \varepsilon_r \) of SNMWO as a function of frequency from 25 to 500 °C is shown in Fig. 2(a). On decreasing frequency, \( \varepsilon_r \) increases and in the low-frequency zone \( \varepsilon_r \) becomes high. The maxima shifts toward a higher temperature side when we increase the frequency (around 200 °C). The polarization (interfacial, ionic, dipolar, atomic and electronic) effect has a profound effect on the \( \varepsilon_r \). The proper nonalignment of the electric dipoles due to the application of an electric field directs the electronic polarization in the sample. Similarly, the loss tangent behaves like \( \varepsilon_r \). The variation in these two dielectric parameters can be explained on the basis of Maxwell and Wagner two-layer model [24]. According to this model, the dielectric behavior of the sample (SNMWO) is high because of the conducting and non-conducting nature of the grain and its surface respectively. During electrical conduction at low frequencies, the activity of electrons increases near the interface; however, the electron activity is reasonably more active at grains in their higher frequency domain. Consequently, the grain boundaries or interface possess more resistance as greater energy will be required to bring the motion of the charges in the sample. Therefore, \( \varepsilon_r \) and tangent loss (insulating parameters) both exhibit high value and which means we need little energy to bring the motilities of the charge. The dielectric loss (tan δ) also decreases with frequency [Fig. 2(b)].

Figure 2(c) illustrates the effect of temperature on \( \varepsilon_r \) at selected frequencies. The \( \varepsilon_r \) value gradually increases as we raise the temperature, and then, it passes through the maxima, for example, at the frequency of 1 kHz we observe the maxima around 100 °C. When the system behaves like a normal ferroelectrics, we get a sharp peak in the plot of \( \varepsilon_r \) versus T. However, the appearance of a broad peak (maximum) in this plot suggests that the material has relaxation characteristics. Figure 2(c) indicates that \( \varepsilon_r \) gradually increases with a rise in temperature, passes through one or two maxima and then decreases. Also, the height of the plot (\( \varepsilon_r \) vs T) decreases with increasing frequency and such typical behavior is referred to as a relax or Ferro-or antiferroelectric material [25]. The Ca or Sr manganite with perovskite structure is a G-type antiferromagnet. However, the doping with lower (Ni) and higher (W) valency atoms at a Mn site in these manganites results in inducing a ferromagnetic phase in the insulating charge ordered manganite matrix. The anomalies observed in the plot of dielectric permittivity with temperature as shown in Fig. 2(c) justifies the induction of small ferromagnetic phase in the present modified manganite (SNMWO) [6]. Furthermore, the anomalies found in the temperature dependence of dielectric permittivity are well known and are typical for ferroelectric transitions, which often occur in two steps, e.g., from paraelectric to ferroelectric and then from ferroelectric to relax or ferroelectric. The increase in dielectric permittivity is too large to be associated with a magnetic transition.

The variation in the loss tangent value with the temperature is given in Fig. 2(d) at selected frequencies. The value of tan δ is minimum at low temperatures and above 300 °C, it increases moderately [Fig. 2(d)]. At higher temperature, there is a small increase in tan δ due to the following reasons: (a) scattering of the charge carriers, (b) defect in the ferroelectric material, (c) oxygen ion vacancies created due to high-temperature sintering of material and (d) superiority of conductivity. Due to all/or some of the above reasons, both the values of insulating parameters of the material decrease with an increase in frequency. This type of investigation on \( \varepsilon_r \) and tan δ helps to manufacture good and promising materials that are used in various energy storage devices.

3.3. Impedance spectral analysis

The electrical characteristics of different materials like ferroelectrics, ionic conductors and other ceramics can be evaluated with the help of a nondestructive CIS: (complex impedance spectroscopy) process. The role of grain, grain-
interface and electrode could be ascertained for SNMWO by this CIS technique. The exact behavior or response of the electronic material could be obtained by simulating the impedance data with the data obtained by related electrical circuits. The response of such multiferroic material with the application of ac current can be well understood by the following formalism [19]. The AC-conductivity ($\sigma_{ac}$) can be determined with the help of the formula: $\sigma_{ac} = \omega \epsilon' \epsilon_o$ where $\omega = 2\pi f$, $\epsilon_o$ = permittivity of vacuum and $\epsilon'$ = permittivity [(relative $\epsilon_r$) having the formalism = $C_p/C_0$, where $C_p$ and $C_0$ are the capacitance of capacitor filled with dielectric and capacitance of identical capacitor in a vacuum, respectively] [26].

The plot of $Z'$ vs. frequency in the temperature range (25°C–500°C) is represented in Fig. 3(a). The real impedance data become less and less when we increase either frequency or temperature. Thus, the conductivity of the material increases. The releasing of the space charge at higher frequencies (> $10^5$ Hz) and at all the temperatures justifies the merger of the curves and decrease in barrier characteristics [27]. The plot of the imaginary part of impedance ($Z''$) vs. frequency of SNMWO at different temperatures is illustrated in Fig. 3(b). Figure 3(b) indicates that the curves are merged at high frequencies and different temperatures which explains the temperature dependency of the relaxation process (electrical). The

![Fig. 2 (a) Variation of $\epsilon_r$ with frequency at selected temperature with inset at 500°C; (b). The effect of frequency on $\tan \delta$ at (a) 25°C and 50°C (100°C–500°C inset), (c) effect of temperature on $\epsilon_r$ at selected frequencies and (d) temperature dependence of $\tan \delta$ at selected frequency](image-url)
The presence of a relaxation phenomenon in SNMWO is verified by these figures [28, 29]. The little broadened peaks in Fig. 3(b) confirms the presence of multiple relaxation times in the material. The reasons for the relaxation can be ascribed to the facts: the movement of the electron under ambient temperature, the creation of Oxygen ion vacancy and the elevated motion of the intrinsic defect. The electrical, impedance and dielectric behavior in the material is due to the combined effect of the hopping of ‘e-,’ Oxygen ion vacancy and possible dislocations [30].

The Nyquist plots for the experimental and model fitted data are represented in Fig. 4 at 25°C and 50°C. The semicircular arcs of these plots suggest the semiconducting nature of SNMWO. The chosen simulated circuit (CQR)(CR) with the necessary components of R, C, Q is given in Fig. 4 (inset), having $C = A(j\omega)^m$ and $Q = A(j\omega)^{n-1}$ being the Jonscher’s universal-components [31, 32]. The simulation of the impedance data is done with the help of the ZSIMP-WIN(Ver-2) code taking the circuit as (CQR)(CR). When we get semi-circular arcs having their centers falling on the real impedance axis, we call the relaxation is of Debye type. Perfect arcs of Fig. 4 suggest that SNMWO is well prepared with a little imperfection. The introduction of the CPE in the RC network accounts for the non-Debye type of relaxation in the material. The constant phase element (CPE) is a diffused layer model which contains both the resistive and capacitive behavior of the material. $Y_{CPE} = X_0(j\omega)^n = M\omega^m + jNo^m$, where $M = X_0 \cos(n\pi/2)$, $N = X_0 \sin(n\pi/2)$, $Y$ being the admittance [31]. And $X_0$ and $m$ are temperature-dependent but frequency-independent physical parameters; $X_0$ is dispersion magnitude and $m$ is frequency exponent. $0 < m < 1$, for the ideal capacitor $m$, is equal to 1 and for the ideal resistor $i$ is equal to 0 [33]. The $R_b$ and $R_{gb}$ data also indicate the semiconducting nature of the SNMWO. Here, the semiconducting trend is followed an NTCR pattern (negative temperature coefficient of resistance) [34]. Table 1 shows the calculated electrical parameters corresponding to the above-stated circuit. Also, the decreasing value of $R_{gb}$ signifies the semiconducting nature of SNMWO. These electrical parameters of SNMWO hint toward possible applications in the electronic devices.

### 3.4. Electrical conductivity

Figure 5(a) shows the variation of ac-conductivity with frequency. It is clearly observed that the ac-conductivity ($\sigma_{ac}$) decreases with the increase in frequency, due to the band conduction, but the conduction due to hopping $\sigma_{ac}$ increases with the frequency. The details about the ac-hopping conductivity were elaborated in the paper by Pollak and Geballe [35]. The total conductivity ($\sigma_t = \sigma_{dc} + B \sigma \omega^{n}$, this relation is referred as Jonscher’s power law: $\sigma_{dc}$ being dc-conductivity, $\sigma(\omega)$ being $\sigma_{ac}$; $B$ is a constant [36]. So, the overall conductivity performance of the SNMWO is due to the band conduction dominance of hopping conduction. The conductivity versus frequency plot obeyed Jonscher’s power law which is shown in Fig. 5(a). The ac-conductivity is calculated using relations; $\sigma_{ac} = \omega\varepsilon_i\varepsilon_0 \tan\delta$ [37]. If we know the Boltzmann constant, ac-conductivity can be determined by the formalism: $\sigma = \sigma_0 e^{-E_a/kT}$. The relation is due to the thermal excitation of electrons from the Fermi level to the space of the maximum density of states. The effect of the temperature $(10^3/T)$ on $\sigma_{ac}$ at selected frequencies is represented in Fig. 5(b). In the high-temperature region, the slope of the plot is invariant and the value of $E_a$ increases with the increase in frequency. $E_a$ values of 1.54, 2.02, 4.67, 9.05 and 13.45 eV at 1 kHz, 10 kHz, 100 kHz, and 500 kHz.
Table 1  Circuit parameters obtained by the (CQR)(CR) circuit d ($C_b$, $Q$, $R_b$, $C_{gb}$ and $R_{gb}$)

| Temp (°C) | $C_b$ (nF/cm$^2$) | $Q$ (S-sec$^{-3}$/cm$^2$) | Frequency power(n) | $R_b$(kΩ·cm$^{-2}$) | $C_{gb}$ (nF/cm$^2$) | $R_{gb}$ (kΩ·cm$^{-2}$) |
|-----------|-------------------|--------------------------|--------------------|-------------------|-------------------|-------------------|
| 25        | 222.3             | 6.80E-09                 | 0.6604             | 354.1             | 1015              | 472.2             |
| 50        | 191.6             | 4.71E-09                 | 0.6705             | 440.9             | 3695              | 127.9             |
| 100       | 173.8             | 2.64E-09                 | 0.7476             | 92.03             | 4264              | 1.388             |
| 150       | 120.3             | 1.41E-09                 | 0.8384             | 18.92             | 3538              | 0.4301            |
| 200       | 25.4              | 1.98E-09                 | 0.8598             | 2.641             | 957.9             | 1.057             |
| 250       | 176.5             | 4.39E-09                 | 0.6442             | 2.002             | 811.1             | 0.08099           |
| 300       | 172.3             | 2.49E-08                 | 0.4951             | 0.9345            | 1.38E-07          | 0.03315           |
| 350       | 144.3             | 3.01E-03                 | 0.01537            | 153.6             | 1031              | 0.119             |
| 400       | 1.88E+21          | 5.12E-09                 | 0.8821             | 63170             | 142.2             | 0.2827            |
| 450       | 2.07E-07          | 1.39E-07                 | 0.6026             | 0.6047            | 52000             | 2.486             |
| 500       | 2.38E-08          | 4.99E-08                 | 0.749              | 10.8              | 235.4             | 0.2227            |

Fig. 5  (a) Plot of ac-conductivity and frequency of SNMW at different temperature.  (b) Variation of $\sigma_{ac}$ with the inverse of absolute temperature ($10^3/T$) at various frequencies
and 1 MHz, respectively. A higher value of $E_a$ can be explained by the pair approximation which obeys perfectly at higher frequencies (one to one hopping among neighbors) [36]. At lower frequencies, the application of pair approximation becomes complex due to the possibility of multiple hopping.

The O$_2^-$ vacancy defect is maximum in the perovskite like SNMWO because it forms a complex structure involving mixed valencies. Also, near these oxide vacancies, a potential well of long-range is created. The O$_2^-$ vacancy and the potential well influence the localization of electrons around the W-atom with the hopping between W$^{5+}$ to W$^{6+}$. In the low temperature, the ac-conduction is due to the motion of the charge carriers in the potential well. The transition metal ions Ni$^{2+}$ and Mn$^{4+}$ can randomly substitute in Sr(Ni$_{1/3}$Mn$_{1/3}$W$_{1/3}$)O$_3$ in the crystal lattice, and the consequences of this substitution are:

(a) Relaxation time associated with the hopping at the potential well related to the $\sigma_{dc}$ of the above power law.

(b) Greater distribution linked to the mobility of the potential well is related to the second term in the power law $\sigma(\omega)$.

Hence, both the relaxations contribute to the overall ac-conductivity.

The supplementary material in Fig. 1-S contains three different XRD patterns of SNMW (a) the sintered SNMW at 1130 °C, (b) powdered sample at 1100 °C and (c) powdered sample at 1000 °C. Figure 2-S contains the plot of ac-conductivity and frequency of SNMW at different temperatures. Figure 3-S shows the temperature dependence of bulk capacitance, bulk resistance, constant phase element, grain boundary capacitance and grain boundary resistance. Figure 4-S shows the UV–Visible DRS optical spectra of the SNMW. Figure 5-S represents the variation in real modulus (M’) with frequency and (b) the variation in imaginary modulus (M’’) with frequency. Figure 6-S represents the Cole–Cole plot of SNMW perovskite.

4. Conclusions

In summary, perovskite SNMWO with monoclinic structure was synthesized successfully by a solid-state reaction route at a moderate temperature. The SEM micrograph shows elongated grains, which are systematically distributed in the SNMWO sample. The sample is exhibited in different thermal relaxation sequences under the hopping mechanism with the charge carriers. The Ca or Sr manganite with perovskite structure is basically a G-type antiferromagnet. However, the doping with lower (Ni) and higher (W) valency atoms at the Mn site in these manganites results in inducing a ferromagnetic phase in the insulating charge ordered manganite matrix. The anomalies observed in the plot of dielectric permittivity with temperature justify the presence of small ferromagnetic phase in a modified manganite. The impedance parameters data are simulated with an equivalent circuit [([CQR]/(CR)), and the Cole–Cole plots were also showed a similar relaxation. Overall, the conductivity versus frequency plot obeyed Jonscher’s power law. The performance of the SNMWO is due to the dominance of hopping conduction rather than the band conduction. With the increase in temperature, the bulk resistance ($R_b$) decreases significantly up to 300 °C; hence, the present material SNMWO exhibits a negative temperature of resistance (NCTR).

Acknowledgments The present work is funded by the UGC-DAE-CSR, Mumbai (CRS-M-297). The authors would like to thank Dr. P. D. Babu of UGC-DAE-CSIR, Mumbai, and Dr. Raja Kishor aLenka of Powder Metallurgy Division, BARC, Mumbai, for providing facilities to conduct.

References

[1] I V Veksler and M P Tepeletev Lithos 26 177 (1990).
[2] C A Triana, L T Corredor, D A L Tellez, and J Roa-Rojas Mater. Res. Bull. 46 2478 (2011).
[3] P A Cox Transition metal oxides: an introduction to their electronic structure and properties (Clarendon Press) (2010).
[4] A Maigian, S Hébert, L Pi, D Pelloquin, C Martin, C Michel, M Hervieu, and B Raveau Cryst. Eng. 5 365 (2002).
[5] A Mleiki, R Hanen, H Rahmouni, N Guermazi, K Khirouni, E K Hill, and A Cheikhrouhou RSC Adv. 8 31755 (2018).
[6] B Raveau, Y M Zhao, C Martin, M Hervieu, and A Maignan J. Solid State Chem. 149 203 (2000).
[7] M B Salamon and M Jaime Rev. Mod. Phys. 73 583 (2001).
[8] C N R Rao and A K Raychaudhuri Colossal magnetoresistance, charge ordering and related properties of manganese oxides, Chap 1, pp 1–42 (2003).
[9] A Urushibara, Y Moritomo, T Arima, A Asamitsu, G Kido and Y Tokura Phys. Rev. B 51, 14103 (1995).
[10] S B Reddy, K Mohan Kant, K P Rao, M Opel, R Gross, and M S R Rao J. Magn. Magn. Mater. 303 e332 (2006).
[11] R Sondenä, P Ravidrann, S Stislén, T Grande, and M Hanfland Phys. Rev. B 74 144102 (2006).
[12] A Aguadero, D Pérez-Coll, J A Alonso, S J Skinner, and J Kilner Chem. Mater. 24 2655 (2012).
[13] T Suzuki, H Sakai, Y Taguchi, and Y Tokura J. Electron. Mater. 41 1559 (2012).
[14] G George, S L Jackson, C Q Luo, D Fong, D Luo, D Hu, J Wen, and Z Luo Ceram. Int. 44 21982 (2018).
[15] S K Dehury, D Khatua, P G R Achary, and R N P Choudhary Indian J. Phys. 93 837 (2019).
[16] J Kim, A Paul, P A Crowell, S J Koester, S S Satapathy, J P Wang, and C H Kim Proc. IEEE 103 106 (2015).
[17] K Parida, S K Dehury, and R N P Choudhary J. Mater. Sci. Mater. Electron. 27 11211 (2016).
[18] S K Dehury, K Parida, and R N P Choudhary J. Mater. Sci. Mater. Electron. 28 10441 (2017).
Studies of structural, dielectric and electrical

[19] K Parida, S K Dehury, and R N P Choudhary Mater. Sci. Eng. B 225 173 (2017).
[20] S K Dehury, P G R Achary, and R N P Choudhary J. Mater. Sci. Mater. Electron. 1 (2017).
[21] P G R Achary, S K Dehury, and R N P Choudhary J. Mater. Sci. Mater. Electron. 1 (2018).
[22] T Chakraborty, H S Nair, H Nhalil, K Ramesh Kumar, A M Strydom, and S Elizabeth J. Phys. Condens. Matter 29 025804 (2017).
[23] T Ramkumar, M Selvakumar, R Vasanthssankar, A S Sathishkumar, and P Narayanasamy J. Magnes. Alloy. 6 390 (2018).
[24] L Alexander and H P Klug J. Appl. Phys. 21 (1950).
[25] A J Dos santos-García, E Solana-Madruga, C Ritter, A Andrade-Chacón, J Sánchez-Benítez, F J Mompean, M García-Hernández, R Sáez-Puche, and R Schmidt Angew. Chemie Int. Ed. 56 4438 (2017).
[26] D Kumari, R Rai, and A Kholkin Influence of BiFeTaO3 addition on the electrical properties of Na 0.4725K0.4725Li0.055NbO3 ceramics system using impedance spectroscopy (2015).
[27] A Kumar, B P Singh, R N P Choudhary, and A K Thakur J. Alloys Compd. 394 292 (2005).

Publisher’s Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.