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Credit Authors Statement

- The corresponding author is responsible for ensuring that the descriptions are accurate and agreed by all authors.
- Rutuparna Das: Synthesis of sample, structural and electrical characterization, prepared the manuscript, R N P Choudhary: Some analysis in the paper and supervise the work.

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Structural and Electrical Properties of Double Perovskite: (BaSr)FeMoO₆

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Abstract:

In this article, the structural, micro-structural, and temperature-frequency dependence of electrical characteristics of double perovskite (BaSr)FeMoO₆ have been reported. The compound crystalizes in cubic symmetry with lattice parameter a=7.9280 (2) Å, and V=245.95 (Å)³. The micro-structural study suggests the uniform distribution of grains on its surface with small voids. The obtained high dielectric constant and low tangent loss value may be applicable for capacitor. In the impedance analysis we observed the behaviour of negative as well as positive temperature co-efficient of resistance. The study of the Nyquist plot suggests the existence of only grain effect. Based on both impedance and modulus spectroscopy, dielectric relaxation process is found to be of a non-Debye type. The frequency dependence of conductivity obeys the Jonscher's Power law which suggests the conduction phenomenon follows the non-overlaping small polaron tunnelling and correlated barrier hopping models. The calculated activation energy in the conduction process may be used in electron hoping. The comparative study of Z'' and M'' with frequency suggests the existence of short range ordering of charge carriers. The leakage current study reveals that the conduction mechanism follows the space charge limited conduction phenomenon.

Key words: Double perovskite; X-ray diffraction; Nyquist plot; Jonscher's power law; Space charge limited conduction; Complex impedance study

1. Introduction:

Now-a-days, there has been growing demand for searching of room temperature magnetoelectric multiferroic materials for both fundamental scientific understanding and to develop the novel multifunctional devices. Multiferroics are the materials in which at least two of the three ferroic properties, namely ferroelectricity, ferromagnetism and ferroelasticity, coexist. Due to the coupling between ferroelectric and magnetic domains, the multiferroic materials offer a novel class of applications in multistage information storage devices, like FeRAM, MRAM etc.[1-2]. Spintronics is a field of investigation for developing the influence of electron spin on the electrical conduction. Spintronic materials possess the unique possibilities for use in new functional microelectronic devices, ideal memory media for computing and in the direction of quantum computing due to its advancement in nonvolatility and magnetic random access memory (MRAM). This technology could also be used to create electronic devices, which should be smaller, faster and consume less power. Hence, the halfmetallic elements with high spin polarization can enhance device performance, and are necessary for a new generation of spintronic devices. There is more attention for the discovery of new perovskite materials having high Tc half-metals. The perovskites (ABO₃) or multiple perovskites belong to a large class of oxide materials have wide industrial applications. The family of double perovskites first received attention in the 1960s [3]. The double perovskite (DP) materials with a general chemical formula $(AA')(BB')O_6$ are the combinations of two perovskites: ABO₃ and A'B'O₃. In (AA')(BB')O₆, A and A' correspond to alkaline or rare earth cations, whereas B and B' are transition metal cations. If A and A' are the similar elements, then the compound has the formula $A_2(BB')O_6$. Such type of materials exhibit wide characteristics, such as semiconducting, conducting, half-metallic, insulating,

ferroelectric, thermoelectric, and superconducting [4-5]. They display interesting magnetic and electronic characteristics with half metallic and low field magneto-resistance. The electric and magnetic properties of the materials are interconnected because of the same (d) character of the "conduction" and "magnetic" electrons. The exchange interaction may happen between ions of the same element along with ions of different elements. The interaction among charge, spin, and lattice results in various electronic and magnetic characteristics, like metallic ferro- or ferri-magnetism or anti-ferromagnetic or paramagnetic insulating property. This also brings about the magneto-resistive properties of the compounds. The electronic and magnetic properties in the double perovskite system can be controlled by substitution of alkaline earth metals (e.g. Ba, Sr, Ca) or lanthanides (e.g. La, Pr, Nd) at the A-site, as well as by the substitution of transition metals (e.g. Fe, Cr, Mn, Mo, Rh, Ru, W, Re). The discovery of low-field magneto-resistance (LFMR) and half-metallic property of the Sr₂FeMoO₆ (SFMO) compound were investigated by Kobayashi et al. [6]. A fully spin polarized half metal, SFMO exhibits a high Curie temperature (>400 K) and excellent magneto-resistance response at relatively small applied fields, and at high temperatures compared to manganites, making it an ideal candidate for room temperature spintronic applications. The magneto-resistive (MR) properties in double perovskite SFMO generally arise from spin-dependent scattering at the grain boundaries. In the double perovskite crystal structure, the oxygen and the large transition metal cations are closely packed, whereas B and B' are arranged inside the oxygen octahedra. The position of the B and B' octahedral is alternating for B and B' elements with a large difference of ionic radii. In the case of a small difference in B and B' ionic radii, a random arrangement of B and B' occurs. The arrangement of the B and B' octahedral influences the B–O–B', B–O–B, or B' – O- B' exchange interactions, and therefore, it is very important for understanding the structural, magnetic, and electronic properties of such type of compounds. The low field

magneto-resistance of such type of compounds is connected to the half-metallic nature of their ground state. The half-metallic nature and the high degree of spin polarization of the charge carriers are also very important for the technical applications of the DP [7-9]. Because of above discussed factors, double perovskite can display a variety of crystal structures for different alkaline and transition metal ions [10-11]. The room temperature structure can be cubic for Sr_2FeMoO_6 [12], Ba_2FeMoO_6 [13]; tetragonal with I4/mmm symmetry, as observed in the case of Sr₂FeMoO₆ [14]; and monoclinic with P2₁/n or P2₁/c: Ca₂FeMoO₆ [13]. The differences between the structures of the same compound, e.g. Sr₂FeMoO₆, can be associated with difference in the chemical composition of DP fabricated under different conditions. As a result, there are some differences in the oxygen stoichiometry along with B, B' ions ordering, and structural defects produced. It is worth noting that when $A_2 = Ba_2$ and SrBa, the compounds crystallize in the cubic structure [15]. Most of the double perovskites have been discussed which show good conducting at a high temperatures with electro-catalytic nature. Sr₂FeTiO₆ is one of the multiferroic double perovskite compounds which are appropriate for sensors and actuators purpose [16]. SFMO (Sr_2FeMoO_6) is a typical double-perovskite (ordered) structure, having an alternating regular arrangement of FeO₆ and MoO₆ in a rock salt super-lattice, where Sr cations make the voids between the octahedral. The radius and charge differences among Fe³⁺ and Mo⁵⁺ are 0.035 and 2 respectively; making fractions of Fe^{3+} and Mo^{5+} reside at the B and B' sites arbitrarily. There is also a fraction of Fe atoms acquiring the B' site, and an equivalent of Mo atoms occupies at the B site which is called the antisite (AS) defect. SFMO has a high Curie temperature (410-450 K) and low-field roomtemperature magneto-resistance. This material may be applicable for magnetic sensors and spintronics owing to its large spin-polarization [17-19]. The properties of double perovskite materials mostly depend on the B-site cations arrangement, which modifies the structural phase transition, electrical and magnetic characteristics. The ordering arrangement can be

controlled by various factors, like the A/B size ratio and oxidation states in the double perovskite materials. For example, dominating characteristics of anti-ferromagnetism of Sr_2LnMoO_6 and ferri-magnetism of CaLaMnMoO₆ has been reported. It is noted that two dissimilar cations at the B-site of double perovskite may permit the two different types of magnetic sub-lattices (a magnetic and a non-magnetic sub-lattice). For instance, the relationship between magnetic 3d cations with non-magnetic 4d or 5d ions leading to ferrimagnetic metallic ordering in Sr₂FeMoO₆, Sr₂CrWO₆, and Sr₂CrReO₆ [20]. From the detailed literature survey on the above double perovskites, it is found that the Fe-Mo containing compounds have recently attracted a great deal of interest of researchers because of their many interesting properties useful for applications. As compared to the manganese based double perovskites, the Fe-Mo materials possess high magnetic ordering temperature which make them suitable for magneto-electronics or spintronic applications. Based on their results and knowing the importance of these compounds, attempt has been made to tailor the physical properties of Sr₂FeMoO₆ by modifying its A-site to get (BaSr)FeMoO₆ compound. Apart from that, some evidences about the magnetic characteristics of such compounds have only been discussed before. Knowing the importance of materials for applications, some interesting work has been reported recently. As the detailed studies of structural and electric characteristics of such material have not been analysed, we have carried out the same (structural, micro-structural, dielectric, and electrical characteristics) of (BaSr) FeMoO₆ which is presented in this paper.

2. Experimental Technique:

The double perovskite ceramic material (BaSr)FeMoO₆ was prepared via a standard ceramic technology solid-state reaction method using high-purity (>99.9%) or AR grade) ingredients, such as strontium carbonate SrCO₃ (M/S Loba Chemie Co Pvt Ltd), barium carbonate BaCO₃

(M/S Central Drug House), iron carbonate FeCO₃ (M/S Central Drug House), molybdenum trioxide MoO₃ (M/S Central Drug House) in stoichiometry. These ingredients were mixed thoroughly in agate mortar and pestle by both dry and wet grinding modes. Then the powder was calcined at 1150[°]C in an alumina crucible. The phase formation was confirmed from the analysis of X-ray diffraction collected by the X-ray diffractometer (Rigaku Ultima-IV) in a wide range of Bragg angle ($20^{\circ} \le 2\theta \le 80^{\circ}$) with a scanning rate of $2^{\circ}/\text{min}$. and CuK_a radiation of wavelength 1.5416 Å. The cylindrical pellets were made by adding polyvinyl alcohol with calcined powder using KBr hydraulic press. The pellets were sintered at 1200°C at atmospheric pressure for 4 h. The room temperature surface morphological study of the sintered pellet was carried out with a scanning electron microscope (SEM, ZEISS). The elemental analysis of the compound was done by studying the energy dispersive analysis of X-ray (EDAX) spectrum. To obtain a smooth surface, two sides of the pellet were polished and coated with high-purity silver paste followed by heating treatment to make moisture-free. By using an impedance analyzer (N4L PSM, 1735), all the electrical parameters were obtained in the frequency range of (1kHz–1MHz) at different temperatures (25–300°C). The leakage current characteristic data of the studied material were obtained using Keithley electrometer (model 6517B).

3. Analysis of results:

3.1 Structural and microstructural analysis:

Fig.1 (a) displays the XRD pattern (room temperature) of (BaSr)FeMoO₆. All the peaks of the X-ray pattern are analysed with the software "Expert Highscore plus". The XRD pattern of this compound consists of cubic (BaSr)FeMoO₆ (ICSD: 01-070-4084) and tetragonal Ba(MoO4) (ICSD: 01-089-4570). The cell parameters were obtained and refined using standard software "POWD MULT" which are: a=7.9280 (2) Å, and V=245.95 (Å)³ (figure in parenthesis implies the observed standard deviation). The crystallite size (D) of the

compound was determined by the Scherrer's equation [21-22]. The magnitude of D is obtained to be 42 nm. Fig.1 (b) displays the room temperature scanning electron microscope of a pellet sample. The grains of different sizes are uniformly (almost) distributed over the surface describing the polycrystalline character of the material. The EDAX pattern (Fig.1(c)) represents a compositional study that shows the existence of essential elements (Ba, Sr, Fe, Mo, O) without any foreign particle. Fig.1 (d) represents the elemental mapping of (BaSr)FeMoO₆ compound.









(d)

Fig.1: (a) X-ray diffraction pattern, (b) SEM, (c) EDAX and (d) elemental mapping of (BaSr)FeMoO₆

3.2 Dielectric Property:

For a ceramic material, the dielectric characteristic is one of the most important properties. Such analysis yields significant information regarding the defects, the quantity of polarization and source of dielectric loss, etc. The dielectric constant of the compound was determined by the relative relation $\varepsilon_r = \frac{C_p}{C_0}$ Where C_p = parallel capacitance (obtained experimentally), C_0

= absolute permittivity [23].



3.2.1 Frequency dependence of dielectric property:



Fig. 2(a-b) is depicted as the frequency dependence of dielectric constant and $\tan \delta$. We observed the systematic and smooth variation of these parameters with temperature and frequency. Since both these parameters reduce with the increment of frequency and temperature, the material behaves like a normal dielectric material. Such a trend in the dielectric spectrum may be ascribed to the various type of polarization (electronic, ionic, dipolar, and space charge polarization). All these polarizations follow the time-varying change electric field in the low-frequency region. Therefore, in such a region, the dielectric constant value is high. All the dipoles may not follow the changing field with an increment of frequency results decrement of dielectric constant [24]. The dielectric dispersion can well be understood by the Koop's phenomenological and Maxwell–Wagner theory [25-26]. According to this theory, the dielectric medium is made up of grains (more conducting) and grain boundaries (less conducting). The grains and grain boundaries are highly sensitive at high and low

frequency respectively. The decrement of ε_r with the enhancement of frequency may be the main cause of hopping of electron at octahedral sites that lags behind the changing electric field. Therefore, electrons pass through both the grains and grain boundaries. Since the grain boundaries are low conducting, the electrons are accumulated near it and develop more space charge polarization. Hence, ε_r value increases in the low-frequency sides. When frequency increases more and more, the electrons suddenly alternate their path, making obstacles for the motion of electrons in the materials, thereby reducing the gathering of electrons and also ε_r . In Fig. 2(b), the dielectric loss shows the similar trend as of dielectric constant. The low-frequency tan δ increases significantly with the rise of temperature representing the part of the dc conductivity owing to the hopping of electrons.

3.2.2 Temperature dependence of dielectric permittivity:

The temperature response of ε_r and tan δ for BSFMO are represented in Fig. 3(a-b). The dielectric constant rises gradually with increasing temperature. Such an increasing trend of the dielectric constant with the enhancement of temperature corresponds to the interaction of electron and phonon [27].



Fig. 3: Temperature dependence of (a) ε_r and (b) tan δ of (BaSr)FeMoO₆

In Fig.3 (a), it is noticed that the room temperature magnitude of ε_r at 1 kHz is nearly equal to 2000, and the value increases with an increase in temperature. Fig. 3(b) displays the variation of temperature dependence of tanð of the studied material. It is observed that at low-temperatures, the magnitude of tanð is low, but the tanð value rises with the enhancement of temperature. This happens because of thermally induced charge carriers and defects. A broad dielectric loss peak is observed corresponding to the quick drop of the dielectric constant. The loss peak shifts to the high temperature side as the frequency increases, suggesting the thermally stimulated dielectric relaxation in the present ceramics. For a pure system, the compound should have a low dielectric loss and high resistance. During calcination and sintering, the charge carriers (electrons) and oxygen vacancies are produced. There may be the chance of the creation of a single/doubly ionized oxygen vacancy with the liberation of one or two electrons from the oxygen. By Kröeger–Vink notation, we have the following equations:

$$O_o = \frac{1}{2} O_2 + V_o'' + 2e^{-}....(2)$$

In this situation, the dipole can be formed by the combination of the ion with a single ionized oxygen vacancy. The dielectric relaxation is affected by these dipoles. The oxygen vacancy is not confined only in the unit cell, but it can affect the total system resulting in the rise of ionic conductivity [28-29]. The stepwise reduction in dielectric constant involves distinct transition, resembles the respective peaks in dielectric loss with the peak positions moves to higher temperatures as frequency raises. This demonstrates the existence of thermally activated relaxation in the material.

3.3 Impedance Analysis:

The complex impedance spectroscopy is usually used to study the electrical characteristics of the materials. The frequency (ω) dependent impedance data are usually analysed in terms of the real and imaginary components of complex impedance (Z), modulus (M), admittance (Y), and permittivity (ϵ). The impedance of the circuit Z(ω) at an applied frequency ω (angular frequency) can be expressed in both polar as well as Cartesian form as below.

$$Z(\omega) = |Z| \exp(-j\theta) = |Z| \cos\theta - j|Z| \sin\theta = Z' - jZ'' \dots (3)$$

Where, Z' and Z'' are the real and imaginary part of the complex impedance (Z) respectively.

For a parallel combination of R (resistance) and C (capacitance) in a circuit, the complex impedance $Z(\omega)$ can be expressed by

$$Z(\omega) = \frac{1}{j\omega c + \frac{1}{R}} = \frac{R - j\omega CR^2}{1 + (\omega CR)^2} = \left[\frac{R}{1 + (\omega RC)^2}\right] - j\left[\frac{\omega R^2 C}{1 + (\omega CR)^2}\right].$$
(4)

After solving the equation, the real and imaginary part of $Z(\omega)$ will be written as

$$\mathbf{Z}' = \frac{R}{1 + (\omega R C)^2} \text{ and } \mathbf{Z}'' = \frac{\omega R^2 C}{1 + (\omega C R)^2} \qquad (5)$$

Admittance: $Y = (Z^*)^{-1}$(6)

Complex modulus
$$M = \frac{1}{\varepsilon^*} = M' + jM''$$
.....(7)



Fig.4: Plot of (a) Z' and (b) Z'' with frequency at different temperature of $(BaSr)FeMoO_6$

Where ω =angular frequency, R= resistance of the material, and relaxation time is determined by using a relation $\tau = R_g C_g$ (C_g, R_g are the effective capacitance and resistance in parallel). Fig.4 (a) and (b) represents the curve of frequency dependence Z' and Z'' respectively. The Z' value decreases at low-frequencies, whereas all the Z' curves merge at a high frequencies. It is indicated that the low-frequency Z' value is temperature-dependent. The reduction of Z' value with the enhancement of temperature (up to 225) indicates the semiconductor type of

behaviour (ie., (negative temperature coefficient of resistance or NTCR)) at high temperatures, whereas for 275⁰C the magnitude of Z' rises with an increment of temperature correspond to PTCR (positive temperature coefficient of resistance) behaviour of the material. The coinciding of high-frequencies Z' value suggests the liberation of space charges leading to the decrease in the activation barrier of the material with a rise in temperature [30-31]. In Fig. 4(b), the magnitude of z'' increases gradually up to a certain frequency. The peak position of the z'' moves in the high-frequency region with the rising of temperature indicating the occurrence of relaxation phenomenon in the compound. The width of the peak determines the distribution of relaxation time. The most probable relaxation time is given by τ_m (=1/ ω_m). The unsymmetrical widening of the peaks demonstrates the occurrence of the electrical process of the sample with a spreading of relaxation times as determined by the peak width [32].



Fig.5: Nyquist Plot of Z' with Z'' (a) at 25-275⁰C, (b) 25⁰C with the depression angle of (BaSr)FeMoO₆

3.4 Nyquist plot:

Fig.5 (a) depicts the Nyquist plots which show correlation between real and imaginary parts of complex impedance. The complex impedance of the materials may be demonstrated by ideal equivalent circuits having resistors R and capacitors C to study the electrical characteristics of the material. The association of the RC network with the constant phase element (Q) indicates the deviation from the Debye-type nature [33-34]. In such a technique, the impedance spectrum is symbolized by semicircular arcs. In this method, the test and theoretical data were verified by (ZSIMP WIN version 2.0) software [35].In the present case, we found a single semicircle (RQC) signifying the occurrence of only grain effect. Apart from that, the depressed semicircular arcs with certain angles (Fig.5 (b)) which centres situated below the real axis corresponding to the non-Debye type of relaxation.

3.5 Analysis of conductivity:

Fig. 6(a) represents the frequency versus ac conductivity curve at various temperatures. This plot consists of a frequency-independent plateau area (dc conductivity) and frequency dispersion region (ac conductivity). The ac conductivity remains frequency-independent up to 100 kHz then the conductivity rises with the enhancement of frequency. The increase of conductivity with the enhancement of temperature may be accredited to free as well as a bound charge from different regions (grain and grain boundaries) in ceramics that are scattered in-homogeneously. This leads to a change in activation energy with temperature [36].To describe the frequency dispersion of the conductivity spectra, the conductivity data can be demonstrated form Jonschor's universal power law [37]

Where σ_{dc} and σ_{ac} are the dc and ac conductivities respectively. The exponent (n) lies in the limit of 0 and 1. The value of n = 1 corresponds to pure Debye case. Fig.6 (b) represents the variation of A and n as a function of temperature. From the curve, it is found that first the value of A increases and then decreases. The behaviour of the exponent factor n as a function of temperature can be used to determine the origin of the conduction mechanism. In CBH (correlated barrier hopping), n reduces with the increase of temperature, whereas, in OLPT (overlapping large polaron tunneling) model, n decreases first, and then increases with increase in temperature. In QMT (quantum mechanical tunneling) mode, the magnitude of n ~ 0.8 and is independent of temperature [38]. In the non-overlapping small polaron tunneling (NSPT) conduction mechanism, the exponent n increases with increasing temperature [39]. Since the magnitude of n increases first, and then decreases, the above discussion of the existence conduction phenomenon belongs to both non-overlapping small polaron conduction (first increasing part) and CBH model (second decreasing part).





Fig. 6: Variation of (a) σ_{ac} with frequency, (b) A and n with temperature and (c) σ_{ac} with 1000/T for (BaSr)FeMoO₆

Temperature (°C)	A	n
25	1.3078E-7	0.00953
75	8.0395E-6	0.00943
125	0.00013	0.01397
175	0.0001	0.02166
225	0.00002	0.02172
275	3.3267E-6	0.02093

Fig.6 (c) represents the variation of ac conductivity with 1000/T of the studied material. Using the dielectric data, σ_{ac} can be determined by the relation:

 $\sigma_{ac} = \omega \varepsilon_r \varepsilon_0 tan \delta....(10)$

Where ε_0 = absolute permittivity. The activation energy can be determined by the expression [40]:

$$\sigma_{ac} = \sigma_0 \exp\left(\frac{-E_a}{K_B T}\right).$$
(11)

Where $k_B =$ Boltzmann constant and $\sigma_0 =$ pre-exponential factor. The ac conductivity of the material increases with the rising temperature that may be the cause of carrier mobility for hopping, which is enhanced by increasing temperature. This behaviour indicates the semiconducting type of material. The values of determining E_a are 0.68, 0.77, 0.79, 0.92, 0.95, 0.91 eV at 1, 5, 10, 100, 500 and 1000 kHz respectively. The merging of high temperature curves suggests the liberation of space charge.

3.6 Modulus Study:

The dynamical nature of the sample can be studied using the complex modulus method. One of the benefits of the modulus formalism is that it significantly suppresses the electrode polarization effects [41]. The advantage of representing the dielectric permittivity data in modulus formalism is that, at lower frequencies, the dominating electrode polarization effects (on dielectric permittivity) are significantly suppressed, and the generally small capacitance values of the grains are enhanced [42]. The frequency variation of the real part of the electric modulus (M') of the studied material is displayed in Fig. 7(a). It is observed that in the low-frequency region, the magnitude of M' tends to zero representing the negligible effect of the electrode, and the M' enhances with the enhancement of frequency. The dispersion of the M' curves with the enhancement of frequency may be the result of the conduction process due to the short-range mobility of charge carriers [43].



Fig.7: Plots of (a) M' and (b) M'' with frequency and (c) M' with M'' and (d) relative analysis curve of M" and Z" with frequency of (BaSr)FeMoO₆

The curves of M" with frequency are depicted in Fig.7 (b). The magnitude of M" rises with the enhancement of frequency, and it exhibits a peak in the high-frequency region. The displacement of M"_{max} peak towards the high-frequency with the enhancement of temperature resembles that the appearance of relaxation processes in the sample. Unsymmetrical widening of the peak suggests the spreading of relaxation with the dissimilar time constant [44].The

Fig. 7 (c) is represented as the variation of the complex modulus spectrum (M' vs. M") of the material. The appearance of a single arc in the spectrum suggests the grain effect in the compounds [45]. Fig. 7 (d) represents a comparative analysis of impedance (Z") and modulus (M"). The matching of Z" and M" peaks resemble the long-range ordering of charge carrier while the mismatching of the peak demonstrates the short-range ordering. In the current study, we observed that mismatching of the peak which suggests the short-range ordering of carrier [46].





Fig. 8(a) Leakage current density (logJ–E) and (b) logJ–logE curve as a function of applied electric fields.

Fig.8 (a) represents temperature variation of current density with electric field (J-E) characteristics of the studied material for both negative and positive bias. The leakage current density was lower than 10^{-3} A/cm² in the presence of +ve and –ve applied of the electric field of 400V/cm which is quite reasonable. There are four mechanisms: Schottky emission (SE), space charge limited conduction (SCLC) mechanisms, modified Schottky– Simmons model,

and bulk-limited Poole–Frenkel (PF)) to understand the conduction process and also to describe carrier transport in ceramics. To know the existence conduction process, the observed J–E data was fitted for quantitative analysis of the charge carrier. First, we have analysed the data for the SCLC mechanism in which the charge carriers are dominated by space charges. The expression for the current density in SCLC is given by [47-48]

$$J_{SCLC} = \frac{9\mu\varepsilon_r\varepsilon_0\theta E^2}{8d} \quad \dots \tag{12}$$

Where, E is the electric field, ε_r is the dielectric constant, ε_0 absolute permittivity, μ is the mobility of carrier, θ is the fraction of injected charge carriers (in trap free limit θ =1). To get information about the conduction process, we have plotted the graph (Fig.8(b)) between logJ and logE. As the obtained slope lies between1-2 so it corresponds to the SCLC mechanism [49-51].

4. Conclusion:

The polycrystalline material (BaSr)FeMoO₆ was successfully prepared by a high-temperature mixed oxide reaction method. The structural study of the material has been carried out by XRD analysis, and the compound is found to be in the cubic system with lattice parameter a=7.9280 (2) Å, and V=245.95 (Å)³. The almost uniform distribution of grains with minor voids is detected by SEM analysis. The EDAX spectrum suggests the existence of the necessary element in the compound without any impurity. The frequency dependence of dielectric study has been explained by the Maxwell-Wagner theory. In the temperature-dependent dielectric characteristics, we have not found any dielectric anomaly. As the room temperature dielectric constant and tangent loss are equal to 2000 and 25 respectively at 1 kHz, the material may be useful for capacitor application. In impedance analysis, both NTCR and PTCR properties of the compound are found. Hence, at low temperatures, this material

may be suitable for semiconductor devices. The frequency variation of conductivity was examined by Jonscher's power law which reveals that the conduction process follows the non-overlapping small polaron tunneling (NSPT) and CBH model. The calculated activation energy in the conduction process may be useful for hopping of electron process. The non-Debye type of relaxation was found from both impedance and modulus study. The occurrence of M" and Z" peaks at different frequency represents short-range ordering. From J-E characteristics, the low value of leakage current density was observed and the conduction mechanism is found to be (SCLC) space charge limited conduction phenomena.

REFERENCE:

[1] S. T. Dadami, S. Matteppanavar, S. I, S. Rayaprol, B. Angadi, B. Sahoo, Investigation on Structural, Mössbauer and Ferroelectric Properties of (1-x) PbFe $_{0.5}$ Nb $_{0.5}$ O₃ – (x) BiFeO₃ Solid Solution, Journal of Magnetism and Magnetic Materials 418, 122 (2016)

[2] S. Matteppanavar, S. Rayaprol, B. Angadi, B. Sahoo, Composition Dependent Room Temperature Structure, Electric and Magnetic Properties in Magnetoelectric Pb(Fe_{1/2}Nb_{1/2})O₃
- Pb(Fe_{2/3}W_{1/3})O₃ Solid-solutions 677, 27 (2016)

[3] N. Kumar, G. Khurana, R. S. Katiyar, A. Gaur, R. K. Kotnala, Double Perovskite Sr₂FeMoO₆: A Potential Candidate for Room Temperature Magnetoresistance Device Applications, doi.org/10.5772/intechopen.70193

[4] C. L. Yuan, S. G. Wang, W. H. Song, T. Yu, J. M. Dai, S. L. Ye, Y. P. Sun, Enhanced intergrain tunneling magnetoresistance in double perovskite Sr₂FeMoO₆ polycrystals with nanometer-scale particles, Appl. Phys. Lett. 75, 3853 (1999)

[5] Y. Tomioka, T. Okuda, Y. Okimoto, R. Kumai, K. I. Kobayashi, Magnetic and electronic properties of a single crystal of ordered double perovskite Sr₂FeMoO₆, Phys. Rev. B 61, 422 (2000)

[6] K. I. Kobayashi, T. Kimura, H. Sawada, K. Terakura, Y. Tokura, Room-temperature magneto-resistance in an oxide material with an ordered double-perovskite structure. Nature. 395, 677(1998)

[7] J. Blasco, C. Ritter, L. Morellon, P. A. Algarabel, J.M. De Teresa, D. Serrate, J. Garcia, M.R. Ibarra, Structural and magnetic details of 3d-element doped Sr₂Fe_{0.75}T_{0.25}MoO₆
Solid State Sci. 4, 651 (2002)

[8] Y. Moritomo, Sh. Xu, A. Machida, T. Akimoto, E. Nishibori, M Takata, M. Sakata, Electronic structure of double-perovskite transition-metal oxides, Phys. Rev. B 61, R7827 (2000)

[9] D. Zaja, M. Sikora, V. Prochazka, M. Borowieca, J. Ste, pien, Cz. Kapusta, P. C. Riedi, C. Marquina, J. M. De Teresa, M. R. Ibarrac, Local Magnetic and Electronic Properties of the A_2 FeM'O₆ (A = Ba, Sr, Ca, M' = Mo, Re) Double Perovskites, Acta Physica Polonica A 111,797 (2007)

[10] F. S. Galasso, F. C. Douglas, R. J. Kasper, Relationship Between Magnetic Curie Points and Cell Sizes of Solid Solutions with the Ordered Perovskite Structure, J. Chem. Phys. 44, 1672 (1966)

[11] N. Auth, G. Jakob, W. Westerburg, C. Ritter, I. Bonn, C. Felser, W. Tremel, Crystal structure and magnetism of the double perovskites A₂FeReO₆ (A=Ca, Sr, Ba),J. Magn. Magn. Mater. 272, 607 (2004)

[12]Y. Moritomo, Sh. Xu, A. Machida, T. Akimoto, E. Nishibori, M. Takata, M. Sakata,Electronic structure of double-perovskite transition-metal oxides, Phys. Rev. B 61, R7827(2000)

[13] R. P. Borges, R. M. Thomas, C. Cullinan, R. Suryanarayanan, L. Ben-Dor, L. Pinsard-Gaudart, A. Revcolevschi, Magnetic properties of the double perovskites A_2FeMoO_6 ; A = Ca, Sr, Ba,J. Phys. Condens. Matter 11, L445 (1999)

[14] J. Navarro, C. Frontera, L. l. Balcells, B. Martinez, J. Fontcuberta, Raising the Curie temperature in Sr_2FeMoO_6 double perovskites by electron doping, Phys. Rev. B 64, 092411 (2001)

[15] C. Ritter, M. R. Ibarra, L. Morellon, J. Blasco, J. Garcia, J.M. De Teresa, Structural and magnetic properties of double perovskites AA'FeMoO₆ (AA' = Ba₂, BaSr, Sr₂ and Ca₂)
J. Phys., Condens. Matter. 12, 8295 (2000)

[16] N. Masta, D. Triyonoa, H. Laysandra, Electrical Properties Sr₂FeTiO₆ Double Perovskite
 Material Synthesized by Sol-gel Method, AIP Conf. Proc. 1862, 030036 (2017)

[17] K. I. Kobayashi, T. Kimura, H. Sawada, K. Terakura , Y. Tokura, Room-temperature magnetoresistance in an oxide material with an ordered double-perovskite structure, Nature 395, 677 (1998)

[18] T. H. Kim, M. Uehara, S.W. Cheong , S. Lee, Large room-temperature intergrain magnetoresistance in double perovskite SrFe_{1-x}(MoorRe)xO₃, Appl. Phys. Lett. 74, 1737 (1999)

[19] J. L. MacManus-Driscoll, A. Sharma, Y. Bugoslavsky, W. Branford, L. F. Cohen, M Wei, Reversible Low \Box Field Magnetoresistance in Sr₂Fe_{2-x} Mo_xO_{6- δ} by Oxygen Cycling and the Role of Excess Mo (x > 1) in Grain \Box Boundary Regions, Adv. Mater. 18, 900 (2006)

[20] S. B. Karki, F. Ramezanipour, Magnetic and electrical properties of BaSrMMoO₆ (M = Mn, Fe, Co, and Ni), Materials Today Chemistry 13, 25 (2019)

[21] N. Pradhani, P. K. Mahapatra, R. N. P. Choudhary, Effect of cerium oxide addition on optical, electrical and dielectric characteristics of (Bi0.5Na0.5)TiO₃ ceramics, J.Phys.: Mater 1, 015007 (2018)

[22] V. Purohit, R. Padhee, R. N. P. Choudhary, Dielectric and Impedance Spectroscopy of Bi (Ca0.5 Ti0.5) O₃ Ceramic Ceramics International. 44, 3993 (2018)

[23] N. Kumar, A. Shukla, R. N. P. Choudhary, Structural, Electrical and Magnetic Properties of (Cd, Ti) Modified BiFeO₃ Physics Letters A 16, 31844 (2017)

[24]] D. K. Pradhan, R. N. P. Choudhary, C. Rinaldi, R. S. Katiyar, Effect of Mn substitution on electrical and magnetic properties of Bi0.9La0.1FeO₃, J. Appl. Phys. 106,024102(2009)

[25] K. Parida, S. K. Dehury, R. N. P. Choudhary, Structural, electrical and magneto-electric characteristics of complex multiferroic perovskite Bi0.5Pb0.5Fe0.5Ce0.5O₃, Journal of Materials Science Materials in Electronics 27, 11211 (2016)

[26] P. Gupta, R. Padhee, P. K. Mahapatra, R. N. P. Choudhary, Structural, dielectric, impedance and modulus spectroscopy of Bi₂NdTiVO₉ ferroelectric ceramics J Mater Sci: Mater Electron 28, 17344 (2017)

[27] N. Shukla, V. Kumar, D. K. Dwivedi, Dependence of dielectric parameters and AC conductivity on frequency and temperature in bulk Se90Cd8In₂ Glassy Alloy, Journal of Non-Oxide Glasses, 8, 47 (2016)

[28] N. Kumar, A. Ghosh, R. N. P. Choudhary, Electrical behavior of Pb(Zr_{0.52}Ti_{0.48})0.5(Fe_{0.5}
 Nb_{0.5})0.5O₃ ceramics, Materials Chemistry and Physics. 130, 381 (2011)

[29] O. Raymond, R. Font, N. Suárez-Almodovar, J. Portelles, J. M. Siqueiros, Frequencytemperature response of ferroelectromagnetic Pb(Fe1/2Nb1/2)O₃ ceramics obtained by different precursors Part I. Structural and thermo-electrical characterization, J.Appl Phys. 97, 084107 (2005)

[30] D. K. Mahato, A. Dutta, T. P. Sinha Impedance spectroscopy analysis of double perovskite Ho₂NiTiO₆, J Mater Sci 45, 6757 (2010)

[31] B. K. Das, T. Das, K. Parashar, S.K.S. Parashar, R. Kumar, H. K. Choudhary, V. B. Khopkar, A.V. Anupama, B. Sahoo, Investigation of structural, morphological and NTCR behaviour of Cu-doped ZnO nanoceramics synthesized by high energy ball milling, Materials Chemistry and Physics 221,419 (2019)

[32] R. Das, R. N. P. Choudhary, Dielectric relaxation and magneto-electric characteristics of lead-free double perovskite: Sm₂NiMnO₆, Journal of Advanced Ceramics 8, 174 (2019)

[33] R. Ranjan, R. Kumar, N. Kumar, B. Behera, R. N. P. Choudhary, Impedance and electric modulus analysis of Sm-modified Pb(Zr0.55Ti0.45)1– $x/4O_3$ ceramics, Journal of Alloys and Compounds 509, 6388 (2011)

[34] S. Hajra, S. Sahoo, R. Das, R. N. P. Choudhary, Structural, Dielectric and Impedance Characteristics of (Bi_{0.5}Na_{0.5})TiO₃-BaTiO₃ Electronic System, Journal of Alloys and Compounds.750, 507 (2018)

[35] S. Sahoo, P. K. Mahapatra, R. N. P. Choudhary, M. L. Nandagoswami, A. Kumar Structural, electrical and magnetic characteristics of improper multiferroic: GdFeO₃, Mater.Res.Express.3,065017 (2016)

[36] T. Das, B. K. Das, K. Parashar, R. Kumar, H. K. Choudhary, A. V. Anupama, B. Sahoo,P. K. Sahoo, S. K. S. Parashar, Effect of Sr-doping on sinterability, morphology, structure,

photocatalytic activity and AC conductivity of ZnO ceramics, J Mater Sci: Mater Electron, DOI 10.1007/s10854-017-7198-6

[37] T. Rhimi, G. Leroy, B. Duponchel, K. Khirouni, S. Guermazi, M. Toumi, Electrical conductivity and dielectric analysis of NaH₂PO₄ compound, Ionics https://doi.org/10.1007/s11581-018-2494-6

[38] S. Kumar, J. Pal, S. Kaur, P. S. Malhi, M. Singh, P. D. Babu, A. Singh, The structural and magnetic properties, non-Debye relaxation and hopping mechanism in PbxNd1–xFeO3 (where x = 0.1, 0.2 and 0.3) solid solutions, Journal of Asian Ceramic Societies 7,133 (2019)

[39] Y. B. Taher, A. Oueslati, N. K. Maalou, K. Khirouni, M. Gargouri, Conductivity study and correlated barrier hopping (CBH) conduction mechanism in diphosphate compound, Appl. Phys. A 120, 1537 (2015)

[40] R. Das, R. N. P. Choudhary, Structure, dielectric and electrical properties of relaxor lead-free double perovskite: Nd₂NiMnO₆, Processing and Application of Ceramics 13, 1(2019)

[41] H. S. Mohanty, A. Kumar, B. Sahoo, P. K. Kurliya, D. K. Pradhan, Impedance spectroscopic study on microwave sintered (1 - x) Na0.5Bi0.5TiO3–x BaTiO3 ceramics, J Mater Sci: Mater Electron, doi.org/10.1007/s10854-018-8683-2

[42] V. Khopkar, B. Sahoo, Low temperature dielectric properties and NTCR behavior of BaFe $_{0.5}$ Nb $_{0.5}O_3$ double perovskite ceramic , Physical Chemistry Chemical Physics 22, 2986 (2020)

[43] R. Das, R. N. P. Choudhary, Structural, electrical, and leakage-current characteristics of double perovskite: Sm₂CoMnO₆, Applied Physics A 125, 864(2019)

[44] S. R. Elliott, Frequency-dependent conductivity in ionic glasses: a possible model. Solid State Ionics 27,131 (1988)

[45] P. Victor, S. Bhattacharyya, S. B. Krupanidhi, Dielectric relaxation in laser ablated polycrystalline ZrTiO₄ thin films, J.Appl.Phys.94, 5135(2003)

[46] D. C. Sinclair, A. R. West, Impedance and modulus spectroscopy of semiconducting BaTiO₃ showing positive temperature coefficient of resistance. J. Appl. Phys. 66, 3850 (1989)
[47] S. Gupta, M. Tomar, V. Gupta, Raman spectroscopy of nanocrystalline Mn-doped BiFeO₃ thin films, Journal of Experimental Nanoscience 8, 261 (2013)

[48] A. Z. Simões, L.S. Cavalcante, F. Mourac, E. Longo, J. A. Varela, Structure, ferroelectric/magnetoelectric properties and leakage current density of (Bi_{0.85}Nd_{0.15})FeO₃ thin films, Journal of Alloys and Compounds. 509, 5326 (2011)

[49] S. Gupta, M. Tomar, V. Gupta, Raman spectroscopy of nanocrystalline Mn-doped BiFeO₃ thin films, Journal of Experimental Nanoscience. 8: 261(2013)

[50] X. G. Tang, J. Wang, Y. W. Zhang, H. L. W. Chan, Leakage current and relaxation characteristics of highly (111)-oriented lead calcium titanate thin films, J. Appl. Phys. 94, 5163(2003)

[51] S. S. Sahu, B. K. Sahoo, A theoretical investigation of pyroelectric effect and thermoelectric improvement of AlInN/GaN heterostructures, Thin Solid Films 684, 59 (2019)

Declaration of interests

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

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: