



Conductivity analysis of $(1-x)\text{Sr}_{0.7}\text{Bi}_{0.2}\text{TiO}_3-x\text{La}(\text{Mg}_{0.5}\text{Zr}_{0.5})\text{O}_3$ ceramic materials

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Abstract

The method of a solid-state mixed oxide reaction was employed to manufacture a strontium bismuth titanate-lanthanum magnesium zirconate ((1-x) SBT- xLMZ) ceramic by a composition of $x = 0.12$ in order to study its structure and conductivity properties. To create the composite, SrCO_3 , Bi_2O_3 , MgO , Ta_2O_5 , ZrO_2 , and TiO_2 were utilized. The ((1-x) SBT-xLMZ) ($x = 0.12$) exhibits a rhombohedral crystal structure, according to the XRD pattern. An XRD was used to measure the X-ray diffraction. The morphological inspection is evaluated using an SEM. An impedance analyzer was used for conductivity measurements. The conductivity at low temperatures is dominated by the mobility of doping-induced extrinsic defects when doping fixes the carrier concentration. Conductivity at high temperatures is caused by thermally produced (intrinsic) defects, whose carrier concentration varies with temperature.

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INTRODUCTION

Ferroelectric materials of perovskite systems are crucial because they are utilized in a wide range of devices, including sensors, transducers, thermally stable ceramic capacitors, and most significantly, the microwave domain (Dhifallah & Turki et al., 2016; Dhifallah & Hehlen et al., 2016). ((1-x) SBT-xLMZ) Being nontoxic, this typical perovskite has exceptional ferroelectric and dielectric characteristics and is also ecologically benign. (Nahar et al., 2020; Chamekh et al., 2020). Most commonly, ((1-x) SBT-xLMZ) is used to replace lead in a variety of applications, such as dynamic random access memory (DRAM) cells. (Nefzi et al., 2013; Said et al., 2014). Due to the high electrical charge storage capacities. To use these materials, we need to understand their electrical properties, such as conductivity. A ceramic

material's conductivity, which is impacted by its microstructure and chemical makeup, is its capacity to carry an electric current. Ceramic conductivity can be either electronic (via electrons) or ionic (via ions), in contrast to metals, which transmit electricity through the movement of free electrons (Kaur et al., 2017).

One important characteristic that sets ceramics apart from metals is the connection between temperature and conductivity. A rise in temperature dramatically boosts conductivity for ionic conductors. This is due to the fact that greater temperatures give ions the thermal energy they need to pass through the material's crystal structure and cross the energy barrier. In a similar vein, semiconducting ceramics' electrical conductivity rises with temperature. This is because when the

temperature rises, more charge carriers, or electrons, are stimulated into the conduction band. This behavior is contrary to that of most metals, where conductivity decreases with rising temperature because of increased electron scattering (Kumar et al., 2017, and Ahad et al., 2019).

Statements of the problem

Ferroelectric materials' electrical conductivity is crucial to study since it affects their pyroelectric, piezoelectric, and other characteristics. A sophisticated method for separating the contributions of the grain and grain border to the total conductivity is impedance spectroscopy. The electric response is obtained across a broad frequency range using the impedance approach. Furthermore, the authors report on their impedance spectroscopy research of ((1-x) SBT- xLMZ) ceramic, which solely shows the grain effect and non-Debye type relaxation (Rayssi et al., 2018). It is rare to find reports on the conductivity analysis of heterovalent substitution on barium strontium titanate ceramics. To the best of our knowledge, the electrical conductivity properties of ((1-x) SBT-xLMZ) (x = 0.12) have not been studied. The aim of this work is to study the conduction mechanism of the solid solution ((1-x) SBT- xLMZ) (x = 0.12). Electrical conductivity as a function of frequency and temperature can be used to better understand the behavior of localized and free electric charge carriers.

Research Questions

1. How does the simultaneous substitution of LMZ at the A-site and at the B-site affect the lattice parameters and the degree of cubic distortion compared to pure SBT?
2. How does the addition of LMZ alter the activation energy for oxygen vacancy migration, and does it successfully suppress the leakage current at high temperatures?
3. To what extent does the LMZ concentration influence the grain boundary mobility of the ceramic?

MATERIALS AND METHODS

The composites ((1-x) SBT- xLMZ) (x = 0.12) are made using the conventional solid-state method. The components that go into creating the composite are SrCO₃, Bi₂O₃, MgO, Ta₂O₅, ZrO₂, and TiO₂. Using an agate motor, all ingredients mixed in a stoichiometric ratio are processed for 16 hours to produce a fine powder. For ten hours, this powdered material is calcined at 900°C in a furnace. The calcined material is ground into a fine powder and mixed with 5% PVC (polyvinyl alcohol) to create a gel. A hydraulic press is used to compress this fine powder into pellets after it has passed through a screen. For 12 hours, this pellet is sintered at 1000°C in a furnace. The following methods were used to characterize the prepared sample. An X-ray diffractometer was used to measure the structure research. A scanning electron microscope (SEM) was used to assess the morphological examination (SEM) and an impedance analyzer was used to measure the conductivity.

Theory

The impedance analyzer was used to measure conductivity. A ceramic material's total electrical conductivity (σ) is the sum of its ionic and electronic conductivity:

$$\sigma_{total} = \sigma_{electronic} + \sigma_{ionic} \quad (1)$$

Although ceramics are typically poor electrical conductors, variables like temperature, composition, and microstructure can greatly change their conductivity. The flow of electrons and holes in ceramics is known as electronic conductivity (σ_e), and it is defined by the following formula (Joshi et al., 2017; Sharma et al., 2015).

$$\sigma_e = n|q|\mu_e + p|q|\mu_h \quad (2)$$

The concentration of free electrons is denoted by N, P is the hole concentration, and the symbol for the elementary charge (1.602×10^{-19} C) is |q|. The symbols μ_e and μ_h stand for electron and hole mobility, respectively.

In ceramics, ionic conductivity (σ_i) refers to the flow of ions across the crystal lattice through lattice defects such as interstitial sites or vacancies. It is

particularly crucial for materials like yttria-stabilized zirconia (YSZ) that are utilized as solid electrolytes. An Arrhenius-type equation that exhibits significant temperature dependence describes this:

$$\sigma_i = \frac{A}{T} \exp\left(-\frac{E_a}{kBT}\right) \quad (3)$$

The pre-exponential factor A correlates with the number of mobile charge carriers and other constants specific to a material. T is the absolute temperature in Kelvin, and E_a is the activation energy of ion movement. The symbol for the Boltzmann constant (1.38 × 10⁻²³ J/K) is kB.

Ionic conductivity rises exponentially with temperature, according to this formula. This is due to the fact that greater temperatures provide ions the energy they need to cross the activation energy barrier and leap to a nearby empty site. Conduction current is created by the movement of charges in the dielectric, which also polarizes the material.

RESULTS AND DISCUSSION

Results

The introduction of LMZ into the SBT matrix induces significant localized structural distortions. X-ray diffraction (XRD) patterns typically reveal a single-phase perovskite structure for low concentrations of LMZ. The system generally maintains a rhombohedral-like symmetry, though Rietveld refinement often indicates slight lattice expansion or contraction depending on the ionic radii mismatch between the dopants and the host ions. Scanning Electron Microscopy (SEM) analysis usually shows that LMZ addition inhibits

excessive grain growth. This leads to a more refined, dense, and uniform grain distribution, which is critical for enhancing dielectric breakdown strength.

The electrical conductivity in these ceramics is governed by both the concentration of oxygen vacancies and the degree of relaxor behavior. The frequency-dependent conductivity typically follows Jonscher's Power Law: In the high-temperature region, activation energy values typically range from 0.23 eV to 0.52 eV, suggesting that the hopping of charge carriers is the primary source of conduction. The activation energy of the compound decreases with increasing frequency. The addition of LMZ generally decreases the overall conductivity compared to pure SBT. This is due to the stabilization of the perovskite lattice and the reduction in the mobility of charge carriers, making the material more suitable for high-energy storage applications.

Discussions

The ((1-x) SBT- xLMZ) (x = 0.12) composite's XRD pattern, observed in the 30–80° range, is displayed in Figure 1. The XRD pattern validates the single-phase rhombohedral structure. It is evident that the composite has a high crystalline nature because Figure 1 shows no further peaks connected to contaminants (Gupta et al.,2018). This material possesses well-crystalline properties, as evidenced by the well-defined powder peak in the XRD pattern of the solid-state reaction.

Table 1

Structural properties of ((1-x) SBT-xLMZ (x = 0.12) ceramic

Composition	Structure	Lattice Parameters (Å°)	Relative Density (%)	Porosity %	Crystallite size (nm)	Average grain size
0.12	rhombohedral	a = 3.54 b = 4.08	96.24	3.76	11.34	1.52

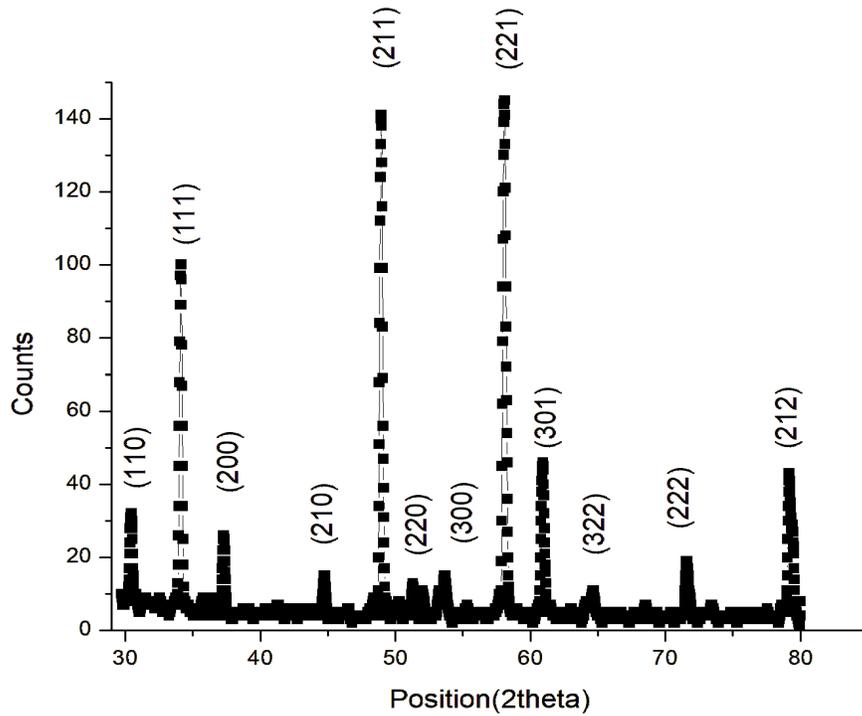


Figure 1. XRD pattern of $(1-x)$ SBT- x LMZ ($x = 0.12$) ceramic materials

Figure 2 displays the micrographs of the composite. This figure shows that the micrograph has a dense microstructure and that the lines dividing two adjacent sections are visible. Table 1 above displays the composite's grain size. The sample appears to have a smooth surface. There are tiny cavities in the sample. Grain boundaries clearly delineate the sample's grain. A straightforward method of

differentiating between contributing elements (grain, grain boundary, electrode, etc.) based on their response characteristics that change with frequency and/or temperature is provided by measurements of Ac conductivity, which also provides insight into a variety of relaxation mechanisms.

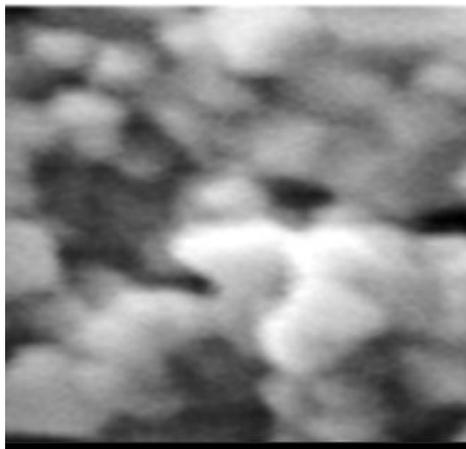


Figure 2. SEM micrographs of $(1-x)$ SBT- x LMZ ($x = 0.12$) ceramic materials
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Distinguish between the contributions of grain and grain border to total electrical conductivity by understanding ac conduction routes. The ac conductivity values are basically calculated using the complex impedance data by:

$$\sigma_{ac} = \frac{e}{S} \left(\frac{Z'}{Z'^2 + Z''^2} \right) \quad (4)$$

Where e is the sample's thickness, and S is the cross-sectional area (Pu et al., 2016).

For x = 0.12, Figure 3 shows the variation in conductivity with temperature at different frequencies. The material's AC conductivity (σ_{ac}) is determined by the sample's dielectric properties. There is a strong correlation between the electrical conductivity of the materials and their frequency response and temperature dependency. The temperature dependence of σ_{ac} indicates the presence of one or more relaxations in the material. The mobility of extrinsic defects, whose carrier concentration is fixed by doping, dominates conductivity at low temperatures. Thermally generated (intrinsic) flaws, whose carrier concentration changes with temperature, are the cause of the conductivity at high temperatures. In contrast to metals, where conductivity declines with rising temperature, the conductivity of many metal oxides increases as the temperature rises. Many metal oxides exhibit an increase in conductivity with increasing temperature. Since the primary factors influencing electrical conductivity in semi-

insulators are crystal defects rather than electrons, the charge carriers in metals, electrical conductivity is a crucial experimental tool for examining structural flaws and internal purity in semi-insulating crystalline solids.

Figure 3 illustrates how the actual part of the ac conductivity varies with 1000/T at various frequencies. The orientation effect is muted and doesn't significantly increase conductivity at lower temperatures, and the complexes that form are stationary. Impurity defect complexes begin to dissolve at a higher temperature, which increases conductivity. Since conductivity values are larger for higher frequencies and exhibit minimal temperature dependency at low temperatures, conductivity is frequency-dependent at low temperatures. The experimental work indicates that the alternative current conductivity (σ_{ac}) increases with temperature and shows negative temperature coefficient of resistance (NTCR) behavior.

In general, the σ_{ac} exhibits an increasing trend as temperature and frequency rise. Impurities or dislocations at the metal-semiconductor interface are responsible for the increase in σ_{ac} with temperature. Since these impurities are situated below the bottom of the conduction band, they have low activation energy. Across a larger range of temperature regions, the type of variation shows essentially linear behavior and adheres to the Arrhenius link.

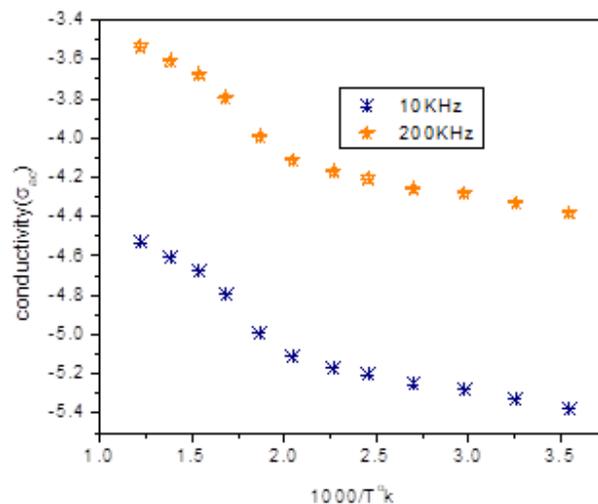


Figure 3. Temperature-dependent variation in conductivity

The high activation energies at high temperatures suggest that the conduction mechanism of the compounds may be due to hopping of charge

carriers. The activation energy of the compound decreases with increasing frequency (Table 2).

Table 2

Temperature and associated activation energies for (1-x) SBT-xLMZ ceramic materials

Composition (x)	Activation Energy (eV)		
	At 10 kHz (250-350°C)	at 300KHz (250-350°C)	At 500 kHz (250-350°C)
0.12	0.52	0.30	0.23

The conductivity variation as a function of frequency plot of the ceramic composition of (1-x) SBT- xLMZ) (x = 0.12) nanopowder is displayed

in Figure 4. There are three distinct regions in a typical conductivity frequency dependency (Hou et al., 2009).

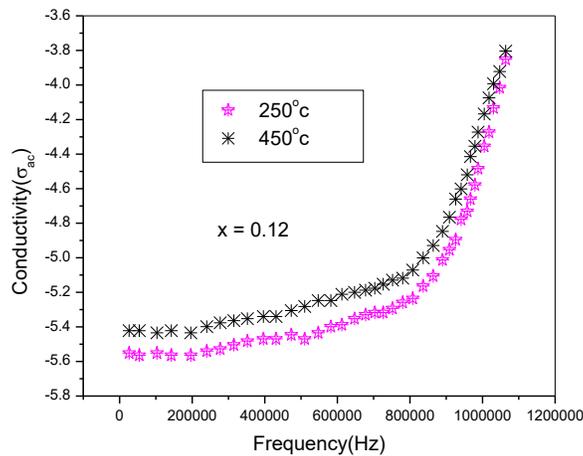


Figure 4. Variation of conductivity as a function of frequency

Low-frequency fluctuations in conductivity are caused by polarization effects at the electrode-electrolyte interface. Due to an increasing quantity of accumulation at the electrode-electrolyte interface, conductivity falls as frequency lowers. Conductivity is nearly frequency independent at the middle frequency plateau, where it equals DC conductivity σ_{dc} . The conductivity in the high-frequency area rises as the frequency does. This figure makes it evident that $\sigma_{ac}(\omega)$ rises as frequency increases.

CONCLUSION

This work effectively produced an electroceramics nanopowder ((1-x) SBT- xLMZ) (x = 0.12) using the solid-state reaction pathway approach and the

precursors SrCO₃, Bi₂O₃, MgO, Ta₂O₅, ZrO₂, and TiO₂. The present work aimed to investigate the structural implications and conductivity property of (1-x) SBT-xLMZ (x = 0.12). This study revealed that the ceramic's crystal structure was rhombohedral. The micrograph's microstructure is thick, making it simple to distinguish the borders between two adjacent sections. The ac conductivity has a propensity to increase with temperature and frequency. The increase in ac conductivity with temperature can be attributed to dislocations or impurities at the metal-semiconductor contact. The electrical relaxation process in the material has been found to be influenced by temperature.

Recommendations

Future reports should include high-resolution TEM to observe polar nanoregions (PNRs), as their interaction with external fields dictates the relaxor behavior and dielectric relaxation in SBT-based systems, and it is recommended to conduct temperature-dependent leakage current measurements to identify the transition from ohmic to space-charge-limited conduction.

CRedit Authorship Contribution Statement

The author confirms they are the sole creator and are responsible for all aspects of this work, including conception, data analysis, and manuscript preparation.

Declaration of Competing Interest

The author declares no conflict of interest.

Ethical approval

Not applicable.

Data Availability

The data generated and interpreted during this research are accessible from the author upon a convincing request.

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