



## Temperature dependence permittivity property study of $(1-x)$ BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> electro ceramic material

Kebede Legesse

Department of Physics, College of Natural and Computational Sciences, Wollega University, Nekemte, Ethiopia

### Abstract

High dielectric permittivity materials could be used in dielectric capacitors because of the demands placed on the integration and shrinking of electronic devices. This study's primary goal was to look into the permittivity and structure of  $(1-X)$  BaTiO<sub>3</sub>– $(X)$  La<sub>2</sub>O<sub>3</sub>. By using a mixture of BaCO<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, and TiO<sub>2</sub> purity of 99.0-99.9 precursors, a twofold sintering solid state reaction was used to create the  $(1-X)$ BaTiO<sub>3</sub>– $(X)$ La<sub>2</sub>O<sub>3</sub> with  $(x=0.18)$  elctro-ceramic nano powder. The sample's structure and permittivity characteristics were ascertained during the characterization procedure using XRD and an impedance analyzer respectively. The crystal structure was determined by XRD examination to be tetragonal, with lattice constants of  $a = 4.52\text{Å}$  and  $c = 5.43\text{Å}$ . The permittivity measurements' results indicate that when temperature rises, the sample's real permittivity constant and imaginary permittivity party first rises peak and then finally fall. Crystal defects form at higher temperatures, which lead to an increase in interfacial polarization. This means that it is predictable for the dielectric constant to rise as temperature does.

### Article Information

#### Article History:

Received: 10-06-2024

Revised : 22-07-2024

Accepted : 30-09-2024

#### Keywords:

Structure,  
Permittivity Properties,  
Batio<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>,  
 $(1-x)$ Batio<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub>

\*Corresponding Author:  
Kebede Legesse

E-mail:  
kebedelegesse@gmail.com

Copyright©2024 STAR Journal, Wollega University. All Rights Reserved.

## INTRODUCTION

Materials classified as dielectrics are those that resist the flow of electric current. Capacitors employ dielectric materials to store electrical charge. Electrical insulation also uses dielectric materials to prevent current from passing through them. Dielectric materials commonly consist of non-metallic materials such as plastics, ceramics, and glasses (Ma et al., 2024; Deng et al., 2023). Barium titanate, a ferroelectric ceramic, has a spontaneous polarization characteristic in

some non-conductive crystals or dielectrics. Capacitors, electromechanical transducers, and nonlinear optics all use this white powder, which is translucent like large crystals. Ferroelectric ceramics are dielectric materials that polarize spontaneously in the absence of an external electric field. Dielectric materials, which are employed as electrically insulating capacitors with an electric dipole structure due to the separation of positive and negative electrically charged entities at the molecular

*Kebede, L.,*

and atomic levels, are extremely poor conductors of electric current (Song et al., 2024; Wang et al., 2024). Because of its many useful applications and exceptional electrical and physical qualities, barium titanate has garnered a tremendous deal of interest throughout the years. Integrated circuits frequently utilize ceramics based on BaTiO<sub>3</sub> for resistors, PTC thermistors, dynamic random access memory (DRAM), and multilayer capacitors (MLCs). Dielectric materials for MLC applications must be electrically insulating, have high permittivity values, and have low dielectric losses at room temperature.

Formulated as BaCO<sub>3</sub>, barium carbonate is an inorganic chemical. Commercial production of barium carbonate involves treating sulfide with sodium carbonate. Like most alkaline earth metal carbonates, it is a white powder salt that is poorly soluble in water and soluble in most acids, with the exception of sulphuric acid. The ceramics industry frequently uses barium carbonate as a glaze ingredient, flux, matting, and crystallizing agent (Liu et al., 2023; Sun et al., 2021). When combined with specific coloring oxides, it can provide distinctive colors that are difficult to achieve with other methods. Art ceramics widely use BaCO<sub>3</sub> to create traditional barium crystal mattes, while BaO crystallizes easily when cooled. Barium carbonate serves not only as an auxiliary material for ceramic coating and optical glass, but also in the production of fireworks and PTC thermistors (Yin et al., 2019; Huang et al., 2019). High-purity barium carbonate is helpful for the electroceramics business, and nanobarium carbonate offers a number of possible uses in science and technology. For

*Sci. Technol. Arts Res. J., July – Sep. 2024, 13(3), 01-07*

solid oxide fuel cathodes, barium carbonate nanoparticles and their mixtures work well as catalysts and are easy to get for a low price. One can determine the PH value of lanthanum oxide by varying the mix of crystal structures. It is a white, odorless, solid that is soluble in diluted acids but insoluble in water. Due to their very desirable characteristics, lanthanum oxide and its metallic oxide are excellent for a wide range of applications, including dielectric materials, catalysts, optical filters, metal supports, and water treatment (Qu et al., 2019). This oxide has an extremely low dielectric constant at E=2, making it the least energy network of choice for both industrial and research labs. The most well-known and extensively utilized application of titanium oxide (TiO<sub>2</sub>) substrates is likely as a bright white pigment and ingredient in sunscreens. TiO<sub>2</sub> substrates have been produced using solid state processes, with sintering temperatures ranging from 1150 to 1350. Because of its strong dielectric and semiconductive qualities, photocatalytic activity, and superior biocompatibility, it has recently garnered more and more attention in the electronics sector (Merkneh & Tadesse, 2020).

The TiO<sub>2</sub> coated thin films are utilized in optical coatings for dielectric mirrors and beam splitters because of its high dielectric constant. It is claimed that the titanium oxide ceramics exhibit both low- and high-temperature crystal formations. TiO<sub>2</sub> ceramics have a wide range of applications, many of which are reliant on the crystal structure. This new research focused on the permittivity qualities of (1-x)BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> (x=0.18) ceramics, taking into consideration the aforementioned properties of the predecessors.

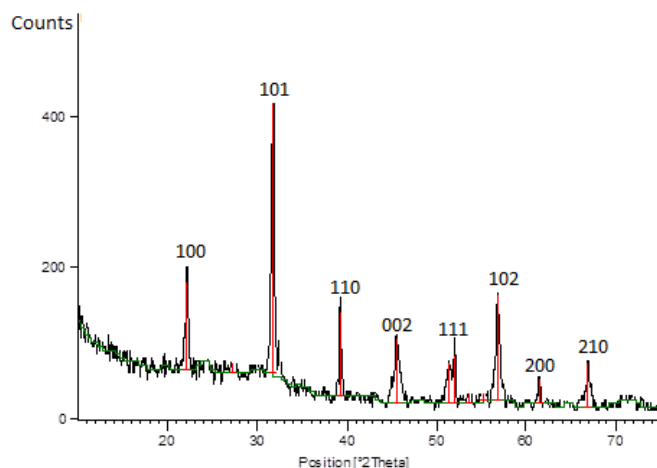
## MATERIALS AND METHODS

(1-x) BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> is the ceramic sample that was examined. Using the solid state reaction technique, (1-x) BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> electro ceramics material was created. The source materials were high purity BaCO<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, and TiO<sub>2</sub>. The sample was made from carbonate precursors and oxides that were of chemical grade (99.0-99.9%). Together with these basic ingredients, the process was aided by acetone (C<sub>3</sub>H<sub>6</sub>O<sub>2</sub>) and powdered polyvinyl alcohol (C<sub>2</sub>H<sub>4</sub>O). Following the sample's synthesis using the chosen method, various measuring meters were used to characterize the sample and determine its properties. Here, X-ray diffraction (XRD) was used to

*Sci. Technol. Arts Res. J., July – Sep. 2024, 13(3), 01-07* characterize the sample's structure, and an impedance analyzer was used to determine the permittivity property of the (1-x)BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> sample.

## RESULTS AND DISCUSSION

To find the crystalline structure of a sample, room temperature X-ray diffraction (XRD) was performed using an X-ray diffractometer in a 2θ range from 10° to 80°. With a lattice parameter of a = 4.52 and c = 5.43, the XRD pattern of (1-x)BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> (x = 0.18), sintered at 950°C for 12 hours, is displayed in Figure 1. There were no extra peaks in the diffraction pattern, indicating pure phase.



**Figure1.** XRD pattern of (1-x)BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> (x = 0.18) ceramics.

Using the Archimede method, the experimental density of the sintered ceramic of (1-x)BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> (x = 0.18) was determined from the specimen that was weighed in both liquid and air. (1-x)BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> (x = 0.18) ceramic had a density of 91.12g/cm<sup>3</sup>. The property of a substance is

influenced by its density in its own unique way. A higher permittivity value is frequently suggested by a higher ceramic density. Therefore, a major factor influencing the difference in permittivity attribute is the density of the substance.

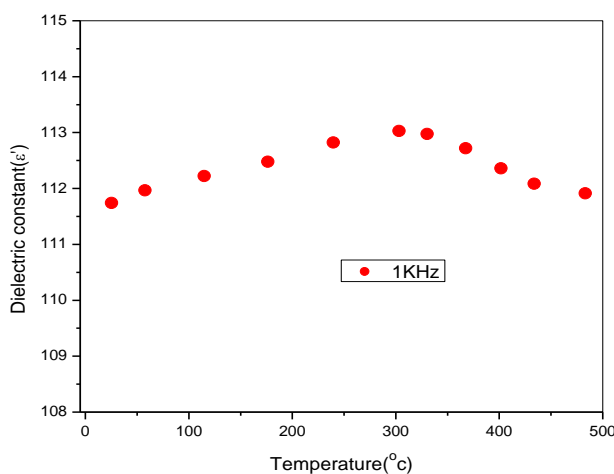
**Table 1**

Structure, crystallite size & density of (1-x) BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub> nanopowder ceramics.

Compositions	Structure	Lattice Parameters (Å <sup>o</sup> )	Relative Density (%)	Porosity %	Crystallite size (nm)	Tolerance factor
0.18	tetragonal structure	a = 4.52 c = 5.43	91.12	8.88 %	22.04	1.060

For the sample (1-x)BaTiO<sub>3</sub>-xLa<sub>2</sub>O<sub>3</sub>, the actual permittivity is plotted as a function of temperature at a constant frequency of 1kHz in Figure (2). The figure illustrates how temperature drastically altered the dielectric characteristics. At 303°C, the Curie temperature, the greatest values of the relative permittivity were found as predicted; however,

as the temperature was raised above this point, the real permittivity dropped to its values at room temperature. The dielectric constant measured in this study at room temperature is 111, which agrees with the values found by (Merkneh & Tadesse, 2020), which indicates that tetragonality and Ferroelectricity which is characterized by a high permittivity constant are connected.



**Figure 2.** Real permittivity ( $\epsilon_r$ ) versus temperature plot at constant frequency.

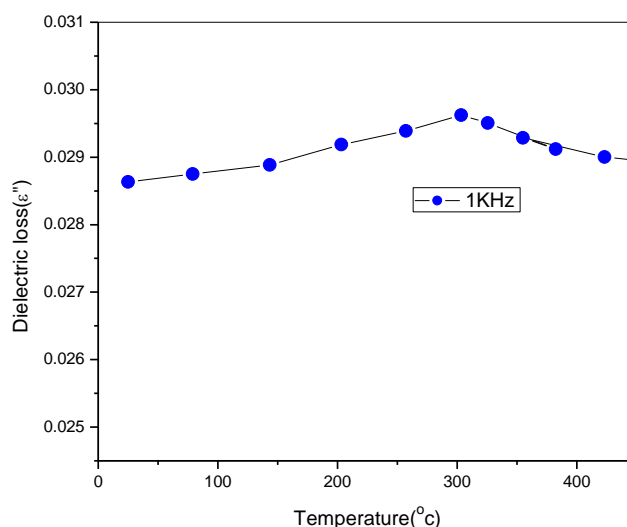
Figure (3) illustrates how the strong faulty structure that gives rise to ions like space

charges is the reason for the increase in dielectric loss. Dipole polarization is the

Kebede, L.,

cause of the increase in dielectric loss at low frequencies (Panda, 2009; Langmuir, 1916). However, at higher frequencies, dielectric loss might exclusively come from ion vibrations ((Ridha & Najim, 2015). For the  $(1-x)\text{BaTiO}_3-x\text{La}_2\text{O}_3$  single crystals, the

*Sci. Technol. Arts Res. J., July – Sep. 2024, 13(3), 01-07*  
temperature dependence of the imaginary component of the dielectric constant was studied at constant frequency beginning at ambient temperature((Petrović et al., 2011;Rayssi et al., 2018).



**Figure 3.** Imaginary permittivity ( $\epsilon''$ ) versus temperature plot at constant frequency.

This graphic illustrates how the imaginary part of the dielectric constant rises noticeably from room temperature to the temperature at which the dielectric constant reaches its maximum at the operating frequency. Nevertheless, the imaginary part of the dielectric constant gradually drops and eventually shows no change after reaching the temperature of maximum dielectric constant.

## CONCLUSIONS

By employing the precursors  $\text{La}_2\text{O}_3$ ,  $\text{BaCO}_3$ , and  $\text{TiO}_2$ , the solid-state reaction pathway technique has successfully produced barium lanthanum titanate  $(1-x)\text{BaTiO}_3-x\text{La}_2\text{O}_3$  ( $x=0.15$ ) electro ceramics nano powder. The present study aimed to produce  $(1-x)\text{BaTiO}_3-$

$x\text{La}_2\text{O}_3$  nano particle powder as an intrinsic dielectric and investigate its dielectric characteristics and structural implications. The ceramic's crystal structures were tetragonal. An increase in temperature at a steady frequency has an impact on the sample's dielectric loss ( $\epsilon''$ ) and dielectric constant ( $\epsilon'$ ).

## ACKNOWLEDGMENTS

The author would like to express his gratitude to Wollega University for providing the necessary supports.

## DECLARATION

The authors declare that they have no conflicts of interest.

Kebede, L.,

## DATA AVAILABILITY STATEMENT

All data are available from the corresponding author upon request.

## REFERENCES

- Deng, C., Li, Y., Wang, H., Qu, Y., Qi, X., Peng, Z., Chen, Z., Shen, H., Sun, K., & Fan, R. (2023). Spark plasma sintered graphene/copper calcium titanate ceramic composites with negative permittivity and enhanced thermal conductivity. *Ceramics International*, 49(10), 16149–16155. <https://doi.org/10.1016/j.ceramint.2023.01.212>
- Huang, X., Yin, R., Qian, L., Zhao, W., Liu, H., Liu, C., Fan, J., Hou, H., Zhang, J., & Guo, Z. (2019). Processing conditions dependent tunable negative permittivity in reduced graphene oxide-alumina nanocomposites. *Ceramics International*, 45(14), 17784–17792. <https://doi.org/10.1016/j.ceramint.2019.05.349>
- Langmuir, I. (1916). The constitution and fundamental properties of solids and liquids. Part i. Solids. *Journal of the American Chemical Society*, 38(11), 2221–2295. <https://doi.org/10.1021/ja02268a002>
- Liu, Y., Cheng, C., Zou, J., Fu, J., Wang, J., Zhou, J., Ma, R., Cui, H., Hu, Z., Wang, T., Du, Y., & Fan, R. (2023). Highly tunable negative permittivity of carbon nanofiber/alumina metacomposites at different external temperatures. *Composites Part a Applied Science and Manufacturing*, 173, 107660. <https://doi.org/10.1016/j.compositesa.2023.107660>
- Ma, R., Cheng, C., Liu, Y., Wang, J., Zhou, J., Hu, Z., Cui, H., Li, J., & Fan, R. (2024). Temperature dependence of negative permittivity behavior in graphene/alumina ceramic metacomposites. *Journal of the European Ceramic Society*, 44(5), 3012–3019. <https://doi.org/10.1016/j.jeurceramsoc.2023.12.016>
- Merkneh, C., & Tadesse, M. (2020). Fabrication and characterization of perovskite ferroelectric BaTiO<sub>3</sub> ceramics from BACO<sub>3</sub> and TiO<sub>2</sub>. *International Journal of Innovations in Engineering Research and Technology*, 7(11), 1–7. <https://repo.ijert.org/index.php/ijert/article/view/52>
- Panda, P. K. (2009). Review: environmental friendly lead-free piezoelectric materials. *Journal of Materials Science*, 44(19), 5049–5062. <https://doi.org/10.1007/s10853-009-3643-0>
- Petrović, M. V., Bobić, J., Ramoška, T., Banys, J., & Stojanović, B. (2011). Electrical properties of lanthanum doped barium titanate ceramics. *Materials Characterization*, 62(10), 1000–1006. <https://doi.org/10.1016/j.matchar.2011.07.013>
- Qu, Y., Du, Y., Fan, G., Xin, J., Liu, Y., Xie, P., You, S., Zhang, Z., Sun, K., & Fan, R. (2019). Low-temperature sintering Graphene/CaCu<sub>3</sub>Ti<sub>4</sub>O<sub>12</sub> nanocomposites with tunable negative permittivity. *Journal of Alloys and Compounds*, 771, 699–710. <https://doi.org/10.1016/j.jallcom.2018.09.049>
- Rayssi, C., ElKossi, S., Dhahri, J., & Khirouni, K. (2018). Frequency and temperature-dependence of dielectric permittivity and electric modulus studies of the solid solution Ca<sub>0.85</sub>Er<sub>0.1</sub>Ti<sub>1-x</sub>Co<sub>4x</sub>/3O<sub>3</sub> (0 ≤ x ≤ 0.1). *RSC Advances*, 8(31), 17139–17150. <https://doi.org/10.1039/c8ra00794b>
- Ridha, S. M. A., & Najim, M. M. (2015). Synthesis and study the dielectric properties of LA-Doped and undoped barium titanate nanopowders. *Engineering and Technology Journal*, 33(2B), 298–306. <https://doi.org/10.30684/etj.33.2b.14>
- Song, X., Shi, G., Fan, G., Liu, Y., & Fan, R. (2023). Tunable Negative Permittivity in Graphene/Poly(Vinylidene Fluoride) Composites with Low Percolation Threshold. *Advanced Engineering Materials*, 26(2). <https://doi.org/10.1002/adem.202300203>
- Sun, Z., Huang, X., Xia, A., Yan, Z., & Qian, L. (2021). Tunable Bandwidth of Negative Permittivity from Graphene-Silicon Carbide Ceramics. *Engineered Science*. <https://doi.org/10.30919/es8d564>

*Kebede, L.,*

Wang, J., Cheng, C., Liu, Y., Zhou, J., Ma, R., Cui, H., Hu, Z., Zou, J., Wang, T., Zhao, Y., & Fan, R. (2024). Tunable negative permittivity performance of carbon/silicon dioxide ceramic metacomposites under external DC bias voltage. *Ceramics International*, 50(5), 7538–7546. <https://doi.org/10.1016/j.ceramint.2023.12.060>

*Sci. Technol. Arts Res. J., July – Sep. 2024, 13(3), 01-07*

Yin, R., Huang, X., & Qian, L. (2019b). Freeze-drying assisted fabrication of highly homogenized reduced graphene oxide/alumina metacomposites with negative permittivity. *Ceramics International*, 45(5), 5653–5659. <https://doi.org/10.1016/j.ceramint.2018.12.030>