CHAPTER 6

Effect of $BaTiO_3$ addition on Electrical, Mechanical and Dielectric Properties of Ceramic Porcelain Insulator

6.1 Introduction

From various articles, we know that alumina electro-porcelain has many advantages, mainly due to it's higher mechanical and electrical strength [Al-Hilli et al. 2010]. Quartz (silica) electro- porcelain is still manufactured because of its lesser price. The significant impact on mechanical properties of quartz porcelain comes from its microstructure where unsolved quartz grains play an important role. It is found that quartz grains (residual) have an adverse effect on the strength of the porcelain described by Liebermann. In quartz porcelain, there is a formation of β cristobalite at a higher temperature, so during cooling mechanical stress comes on insulator due to β-α quartz transformation which occurs at 573°C. It was also noted that the large size of quartz particles results in a high density of microcracks [Štubňa et al. 2007, Chmelík et al. 2011 and Islam et al. 2004]. On the other hand, the effect of addition of fine-grained quartz powder increases the bending strength of porcelain because sub-micron particles dissolve into a feldspatic liquid phase. Both mechanical strength and Young's modulus increase if the quartz content decreases [Magagnin et al. 2014]. This problem resolve by adding zirconia concentration on alumina-silica based porcelain insulator (chapter 4). On addition of zirconia, there is the formation of zircon phase on sintering temperature above 1250°C and decrease the β cristobalite phase of silica [Verkerk et al. 1982, Martin et al. 2003 and Mehta et al. 2018]. In chapter 6, barium titanate ($BaTiO_3$) is used as a dopant in the base composition of CPI for improving the electrical properties of the resultant material. As know that $BaTiO_3$ is

perovskite material and having wide application in the ceramics and electronics industries [Nayak et al. 2014, Gromada et al. 2017 and Abdelal et al. 2014]. $BaTiO_3$ used as a dielectric material due to its high dielectric constant. The mechanical properties of $BaTiO_3$ have less attracted. [De with, 1993]. The mechanical properties depend strongly on the micro- structure [Tuan et al. 1994 and Chang et al. 2011]. $BaTiO_3$ are fabricated at high temperatures using solid-state reaction techniques [Osman 2011].

In this article, the effect of $BaTiO_3$ (0 to 2 wt. %) addition on the physical, the mechanical and dielectric strength of prepared porcelain insulator were investigated. Prepared samples were sintered at 1350°C.

6.2 Material and Experimental Characterization

We have already discussed the base material of ceramic porcelain insulator (CPI) in our previous chapters. $BaTiO_3$ (0 to 2 wt. %) is used as a doping material in the base composition of CPI and done variation between ZrO_2 and $BaTiO_3$. The composition with different code (E1, E2, E3, and E4) of the ceramic porcelain insulator is shown in

Table 6. **1**. The detailed procedure for manufacturing of CPI is already discussed in chapter 2 (section 2.1 and 2.2) and previous chapters.

Prepared compositions powder is compressed by a hydraulic press machine (applying load of 160 MPa) for making testing samples, having different shapes and dimension shown in Figure 6. 1.

Table 6. 1: Variation of ZrO_2 and $BaTiO_3$ in the base composition.

Samples	Ball clay	Kaolin	Feldspar	SiO_2	Al_2O_3	ZrO_2	$BaTiO_3$
E1	20	25	10	10	27.5	7.5	0
E2	20	25	10	10	27.5	7	0.5
E3	20	25	10	10	27.5	6.5	1
E4	20	25	10	10	27.5	5.5	2

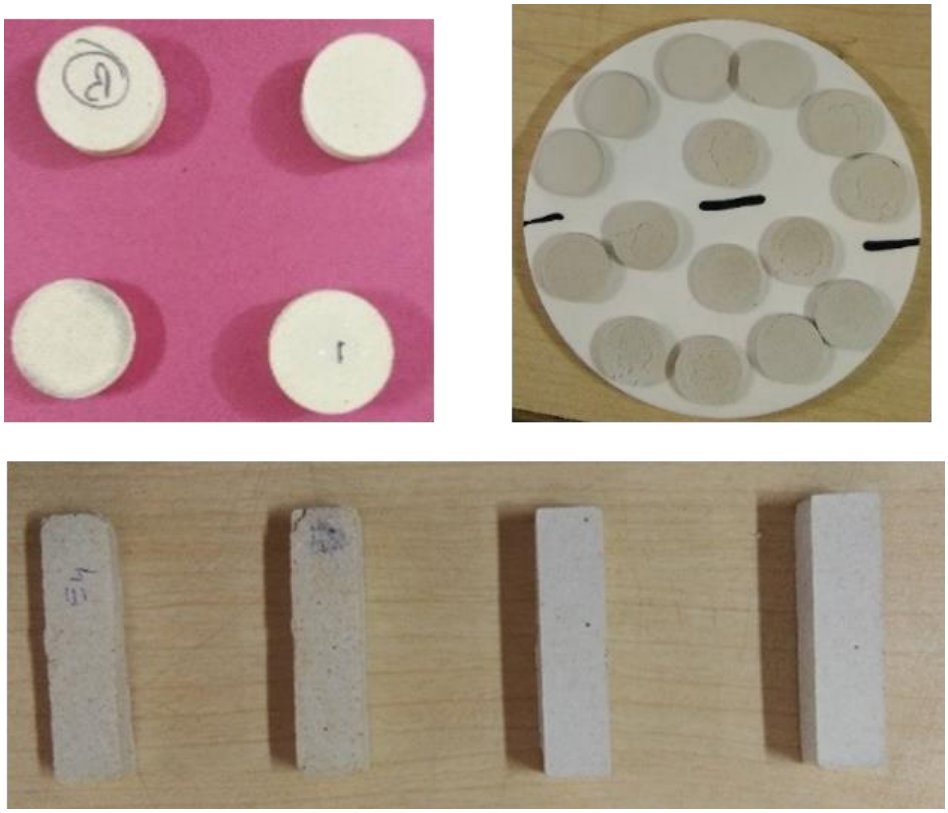


Figure 6. 1: Prepared samples of different compositions sintered at 1350°C.

Further, these prepared pallets or samples sintered at 1350°C with a sintering rate (5 °C/minute) and soaking time are 2 hours at peak temperature (1350°C). Different shapes

(rectangular, square and circular) of samples were prepared for different characterization, i.e., physical, electrical and mechanical testing of porcelain insulator.

The thermal expansion coefficient behavior of different sample (E1, E2, E3 and E4) of porcelain insulator were investigated by dilatometer (from room temperature to 1300°C) with a heating rate of 5 °C/minute. Alumina was used as the reference material in dilatometer. The phase structure of porcelain insulator was performed using the X-ray diffraction analysis with the rate of 5°C in the 2θ range from 10 to 80°. The scanning electron microscope (SEM) and EDS were used for observing the microstructures and element composition of the prepared CPI sintered samples.

6.2.1 Physical Properties

Physical properties such as bulk density, apparent porosity and water absorption of the different composition sintered samples were measured by Archimedes method. The samples were kept in hot water bath (at temperature 80°C) for 5 hours. Measured the wet and soaking weight of different samples and calculate all physical parameters.

6.2.2 Mechanical Characterization

For mechanical characterization measurement using UTM (Universal Testing Machine) such as modulus of rupture (MOR) or bending and compressive strength of the sintered samples at room temperature. The detailed procedure for mechanical testing analysis is discussed in chapter 2.

6.2.3 Electrical Characterization

For electrical characterization such as AC dielectric strength was measured using high voltage dielectric break down voltage tester (NPTL Neo Tele Tronix Pvt. Ltd. Kolkata). The test set up consists of two spherical electrodes with a diameter of 10 mm. The thickness of the sample was kept between 1.2 mm to 1.5 mm, and having a diameter of 15mm is inserted between two spherical electrodes. The voltage applied across the specimen is raised gradually (at the rate of 1kV/s) till sample fails. To avoid any surface flashover, the sample electrode assembly is submerged in transformer oil, the test was followed by the [ASTM standard D149-97a, (2007)].

The AC dielectric and dielectric loss or loss tangent (tanδ) properties of different sintered samples were measured at a frequency range of 1–20 GHz using vector network analyzer. Also to measure this AC electrical characterization at a low-frequency range (20 Hz to 1 MHz) using impedance network analyzer.

6.3 Result and Discussion

6.3.1 Morphological Analysis

The XRD patterns of different samples are having $BaTiO_3$ (BT) content (0, 0.5, 1 and 2 wt. %) and sintered at 1350°C are shown in Figure 6. 2. The analysis results show that the samples are mainly composed of corundum (PDF 46-1212), mullite (PDF 79-1456), m-ZrO_2 (PDF 65-1024), zircon (PDF 83-1374), barium aluminum titanate (PDF 78-1017), cristobalite (PDF 39-1425) as well as amorphous phase. As the concentration of $BaTiO_3$ increased, and zirconia decreased, the major peaks intensity enlarged at an angle (2θ) of 25.7 and 28.2° corresponding to barium aluminum titanate (BAT) because of the dissolution of alumina into barium titanate concentration at high temperature. At the same time, there is a little enhance in the intensity peaks (m-ZrO_2 and zircon) of the samples in

E1 and E2 after that its phase intensity decrease by little shown in Figure 6. 2. The formation of zircon is due to the solid-state reaction between tetragonal zirconia and silica (amorphous and cristobalite).

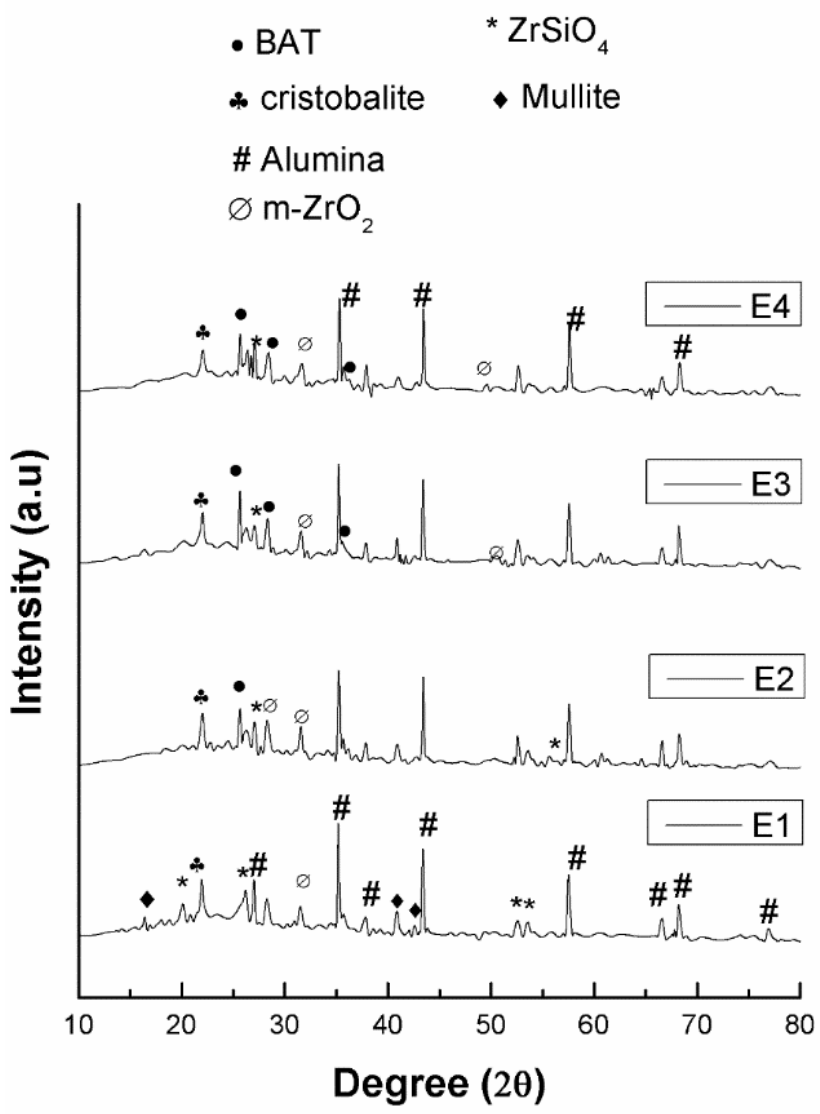
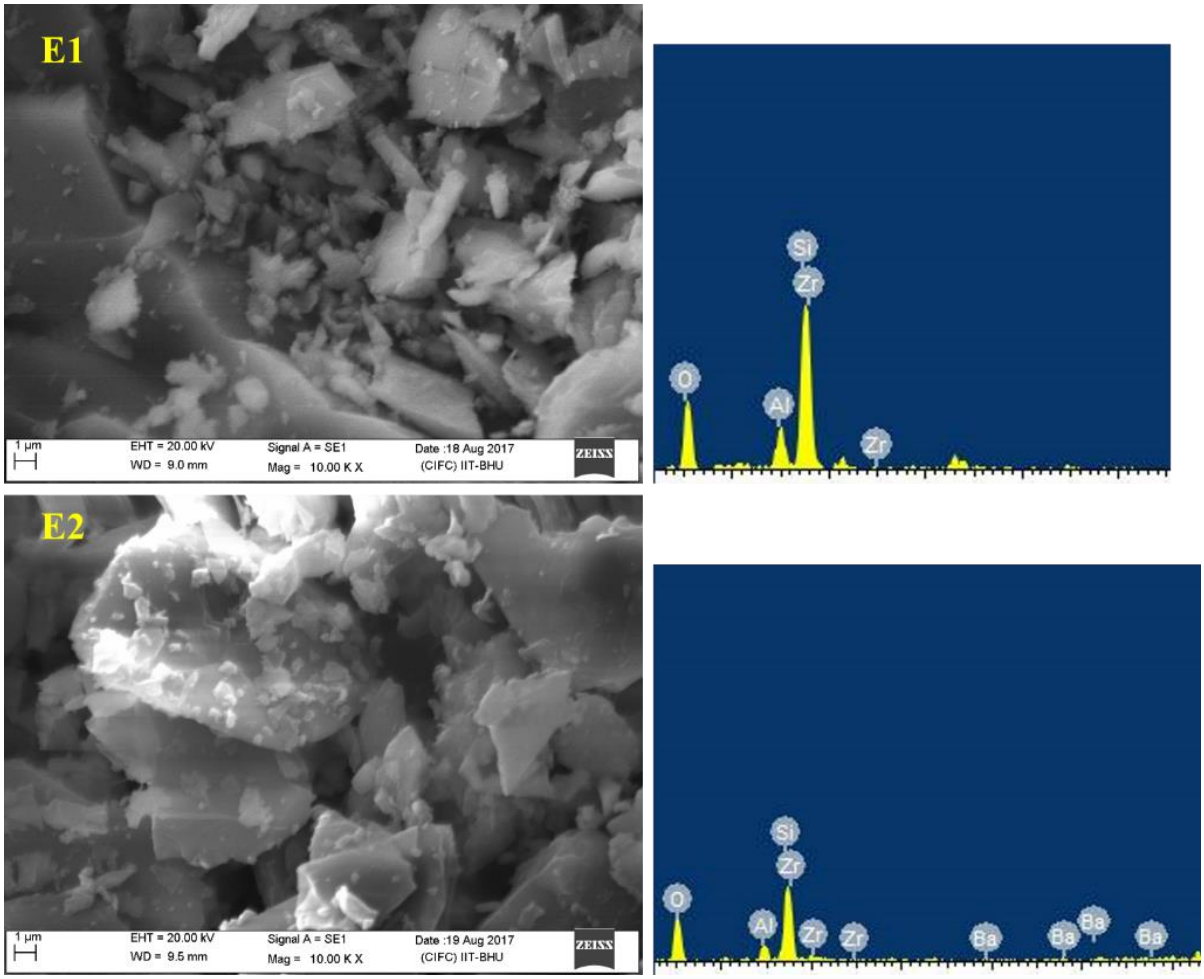


Figure 6. 2: XRD pattern of samples (E1, E2, E3, and E4) sintered at 1350°C; #: alumina, ♦: mullite; ♣: cristobalite; *: $ZrSiO_4$; ∅: m-ZrO_2 and •: BAT.

Figure 6. 3, Shows the surface morphology of four different compositions sintered at 1350°C are analyzed using SEM and EDS. The surface topographies confirmed the presence of little agglomeration and porosity in the samples are present. For the property of good insulators pores, shapes and particle size play a very important role. From study

conform that samples contain various shape such as circular, elliptical and irregular, and homogeneously distributed in the body. The porosity of all four different compositions having BT (0 to 2 wt. %) concentration is measured by Archimedes method are listed in Table 6. 2.



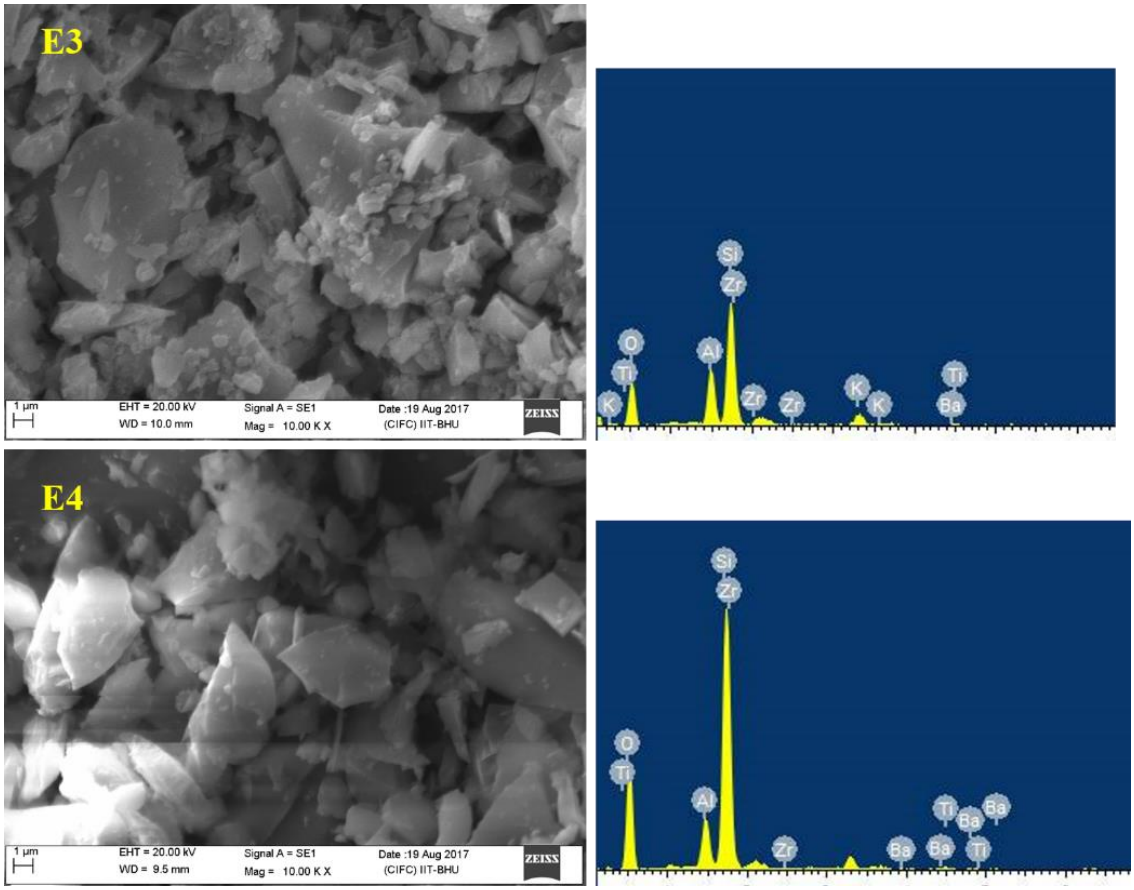


Figure 6. 3: SEM micrograph of the different samples (E1, E2, E3 and E4) was investigated.

The coefficient thermal expansion of the four different sample was examined in the temperature range (30 °C–1300 °C) using dilatometer with heating rate of 5°C/minute. For all samples, shows negative expansion up to approx. 180°C followed by a positive expansion up to 1200 °C. Each samples showed thermal expansion coefficient in the range of 10^{-6}. As we know that BT is more insulating as well as having good mechanical and thermally resistant material. From Figure 6. 4, it reveals that the thermal expansion coefficient of the prepared composition increases significantly as we increase the concentration of BT concentration with reducing zirconia content.

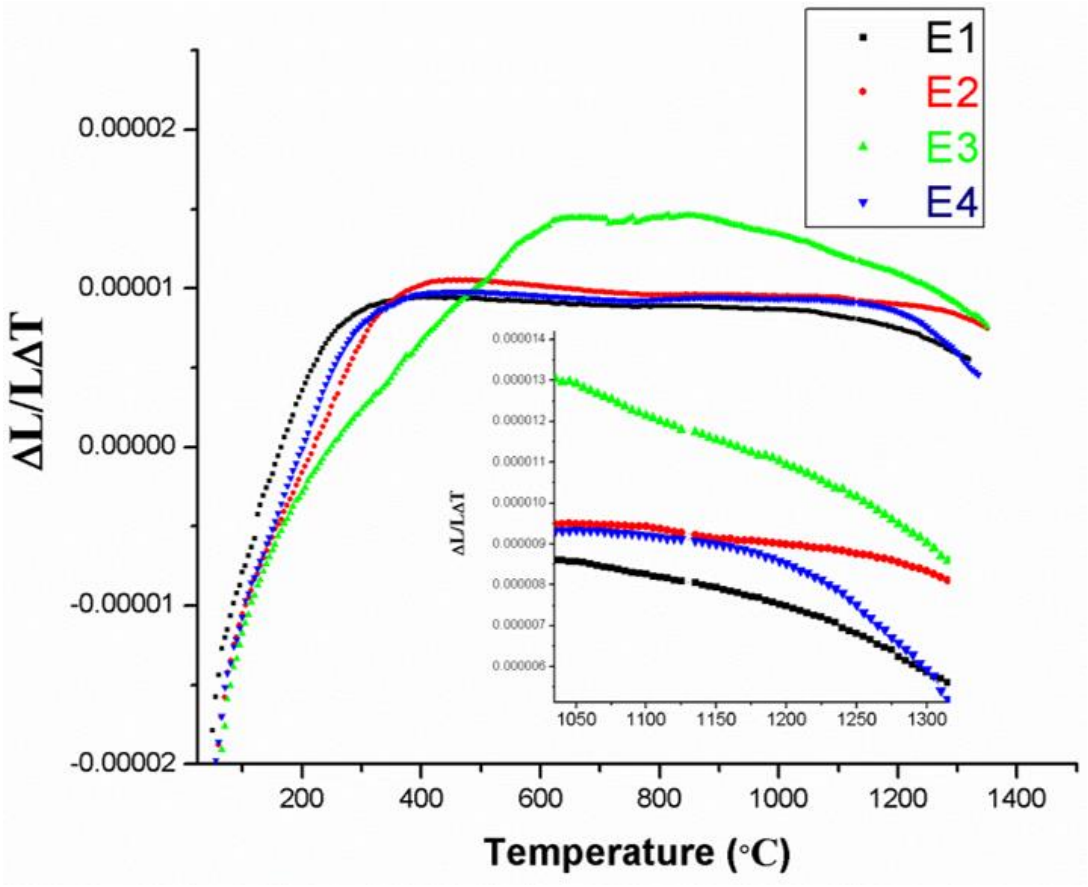
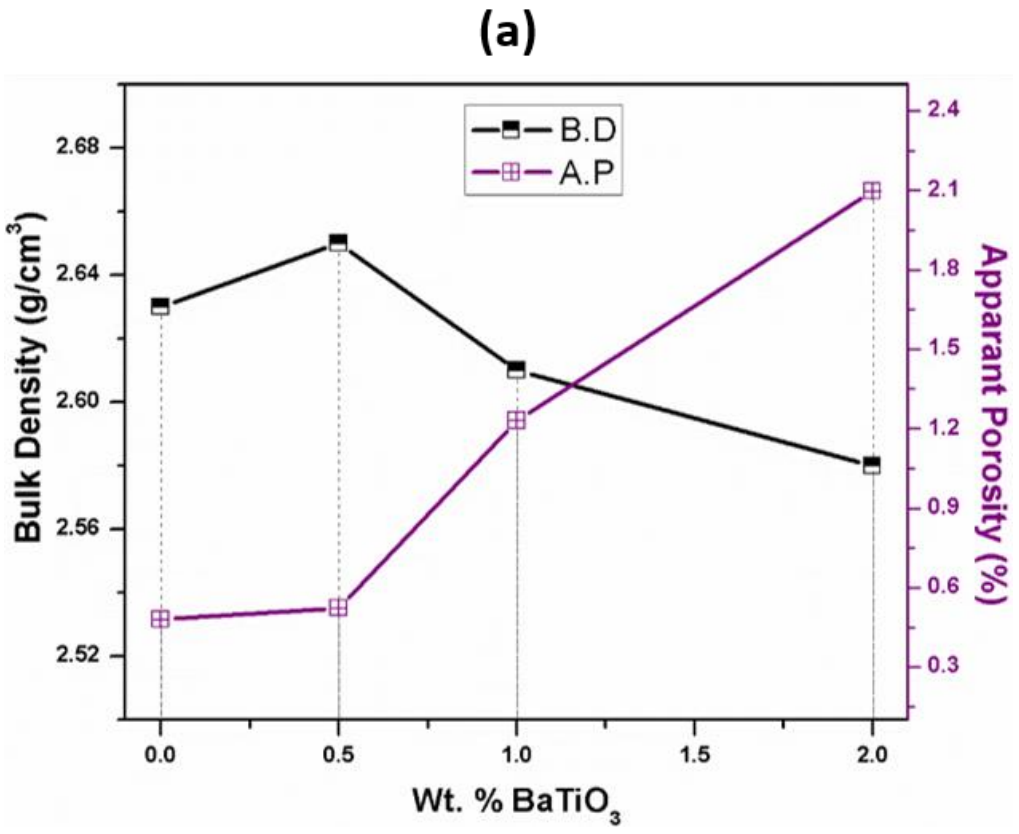
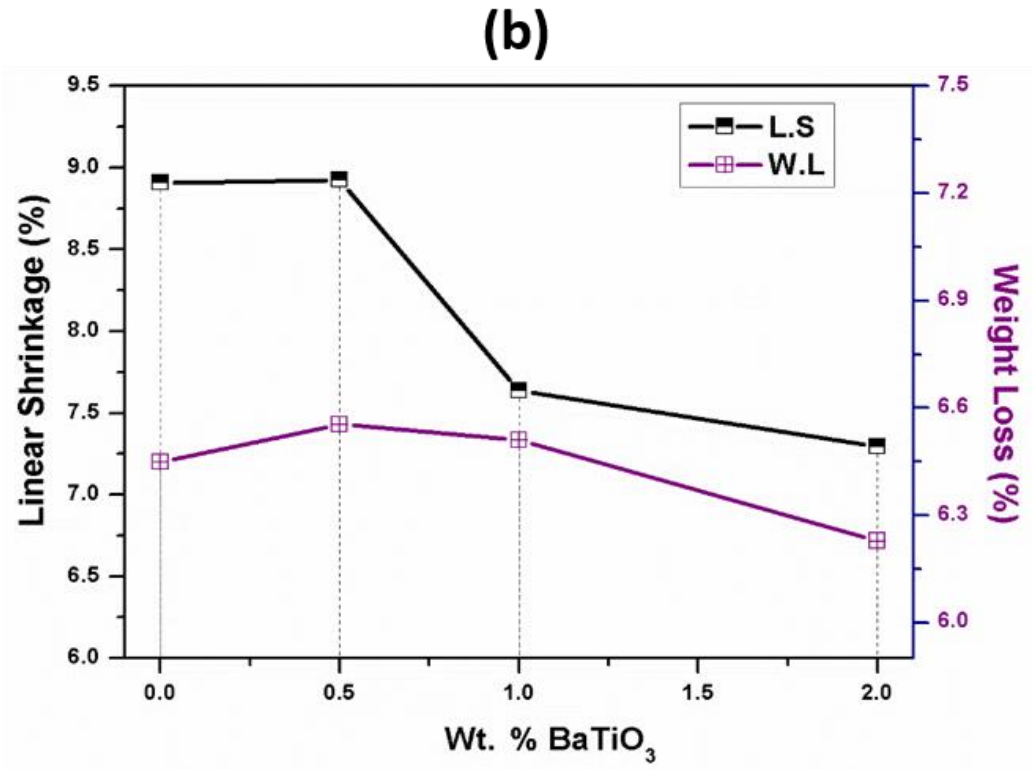


Figure 6. 4: Variation of the coefficient of thermal expansion with temperature after addition of $BaTiO_3$ (0 to 2 wt. %) content in porcelain composition.

6.3.2 Physical Analysis

Figure 6. 5, shows the bulk density (g/cm^3) and apparent porosity (in %) values of four different samples sintered at 1350°C. The samples E1 and E2 showed increased densification on the addition of BT up to 0.5 w.t % in base composition. Futher, addition of BT (0.5 wt. %) on base composition the densification of the sample increases due to increment in porosity, shown in Figure 6. 5 (a).


(a)
B.D
A.P
Bulk Density (g/cm3)
Apparant Porosity (%)
Wt. % BaTiO3
2.68
2.64
2.60
2.56
2.52
2.4
2.1
1.8
1.5
1.2
0.9
0.6
0.3
0.0
0.5
1.0
1.5
2.0


(b)
L.S
W.L
Linear Shrinkage (%)
Weight Loss (%)
Wt. % BaTiO3
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
7.5
7.2
6.9
6.6
6.3
6.0
0.0
0.5
1.0
1.5
2.0

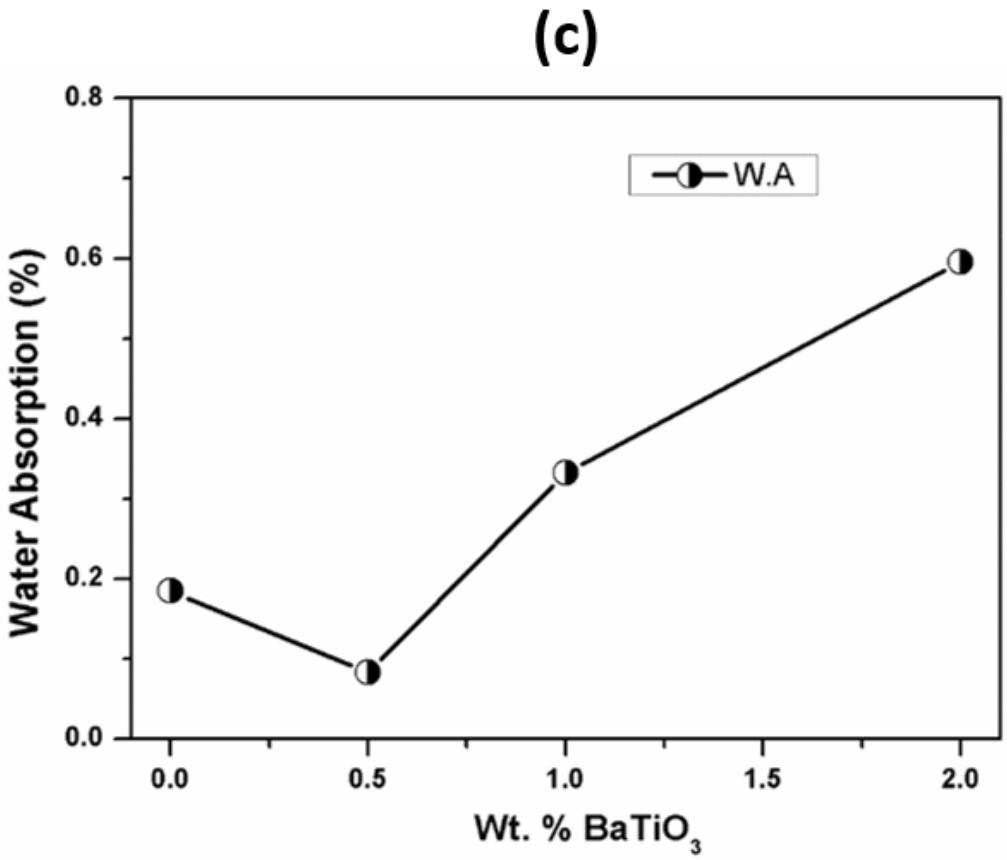


Figure 6. 5: Physical behavior testing of the different sintered samples at 1350 °C.

This densification proceeded due to the grain boundary diffusion and also densification results were confirmed by measuring the linear shrinkage (L.S) of the compacts samples sintered at 1350°C, shown in Figure 6. 5 (b).

The water absorption (W.A) of samples decreased with increasing BT addition up to 0.5 wt. %, it is almost near to zero. For high voltage electrical ceramic porcelain insulator water absorption should be minimal or zero, its leads the electrical resistance properties of the materials. From Figure 6. *5* (a and c), it depicts that W.A and A.P dropped to a minimum by addition of 0.5 wt. % BT with 7 wt. % ZrO_2 in the base porcelain composition sintered at 1350°C. The maximum bulk density (2.65 g/cm^3) with also

maximum linear shrinkage (8.92 in %) were obtained for above said composition (E2), shown in Table 6. 2.

Table 6. 2: The B.D (in g/cc), A.P (in %), L.S (in %), W.L (in %) and W.A (in %) with content of BT (0–2 wt. %) sintered at 1350°C with soaking period for 2 hours.

Samples sintering at 1350°C				
Wt.% $BaTiO_3$	**E1**	**E2**	**E3**	**E4**
B.D (g/cm-3)	2.63	2.65	2.61	2.58
A.P %	0.47	0.52	1.22	2.09
L.S %	8.90	8.92	7.63	7.29
W.L	6.44	6.55	6.51	6.22
W.A %	0.18	0.08	0.33	0.59

6.3.3 Mechanical Characterization

The maximum observed mechanical strength in samples (E2) such as the bending, tensile and compressive strength values of porcelains sintered at 1350 °C for 2 hours were 2.65 g/cm^3, 144 ± 5 MPa, 40.50 ±3 MPa and 223 ± 10 MPa, respectively shown in Figure 6. 5, Figure 6. 6 and Figure 6. 7. After further addition of BT (up to 2 wt. %) content, i.e. samples E3 and E4, its physical and mechanical strength goes to decline, due to leads in porosity on that samples, shown in Table 6. 2 and Table 6. 3.

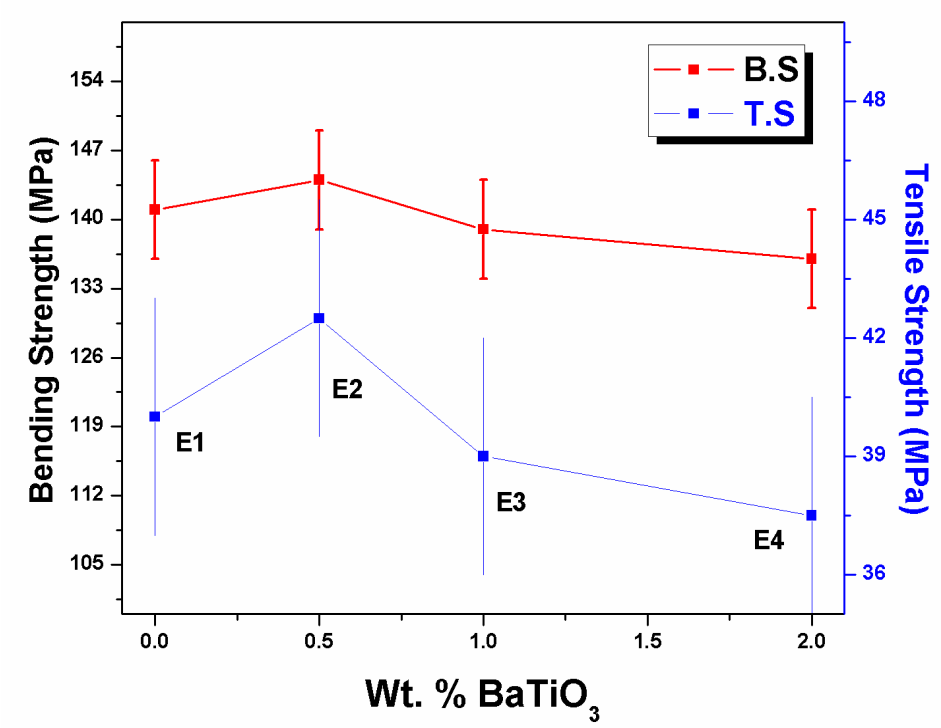


Figure 6. 6: Graph between B.S, T.S (in MPa) versus BaTiO3 wt. %, for the samples sintered at 1350°C.

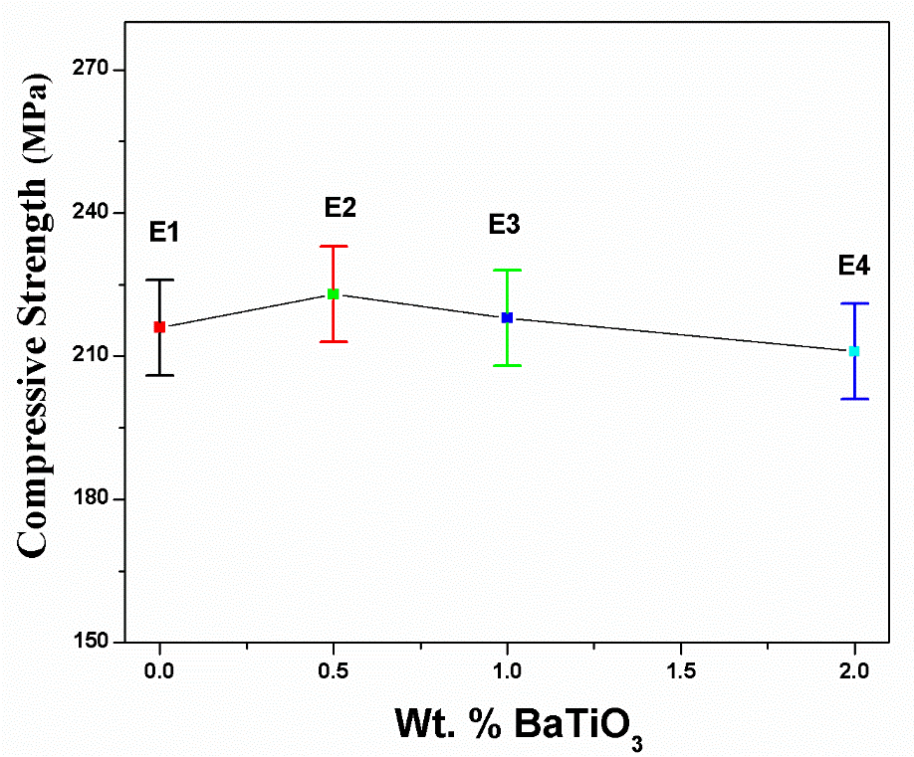


Figure 6. 7: Graph of C.S (in MPa) versus $BaTiO_3$ wt. %, for the samples sintered at 1350°C.

Due to the change in the stress state, the deformation behaviors of the composites are strongly influenced by their local stress tension or compression. That's the reason that the compressive properties are always higher than the tensile and bending properties.

Table 6. 3: The calculated value of Modulus of rupture (MOR) and compressive strength of different samples with temperatures.

Samples	$BaTiO_3$ Contents (Wt. %)	MOR (MPa)	Compressive Strength (MPa)	Tensile Strength (MPa)
		1350 °C		
E1	0	141 ± 5	216 ± 5	40.00 ± 3
E2	0.5	144 ± 5	223 ± 5	40.50 ± 3
E3	1	139 ± 5	218 ± 5	39.00 ± 3
E4	2	136 ± 5	211 ± 5	37.50 ± 3

6.3.4 Electrical Characterization

The dielectric loss represents a combined result of electrical conduction and orientation polarization of the matter. Shows in Figure 6. 8 (b) the variation of dielectric loss tangent (tan δ) with frequency for different sintered samples. The investigated composition shows a decreases in dielectric behaviour and Dielectric loss with increasing frequency (from 20 Hz to 1 MHz), shown in Figure 6. 8. The values of tan δ decrease at higher frequency in the range 0.01–0.009.

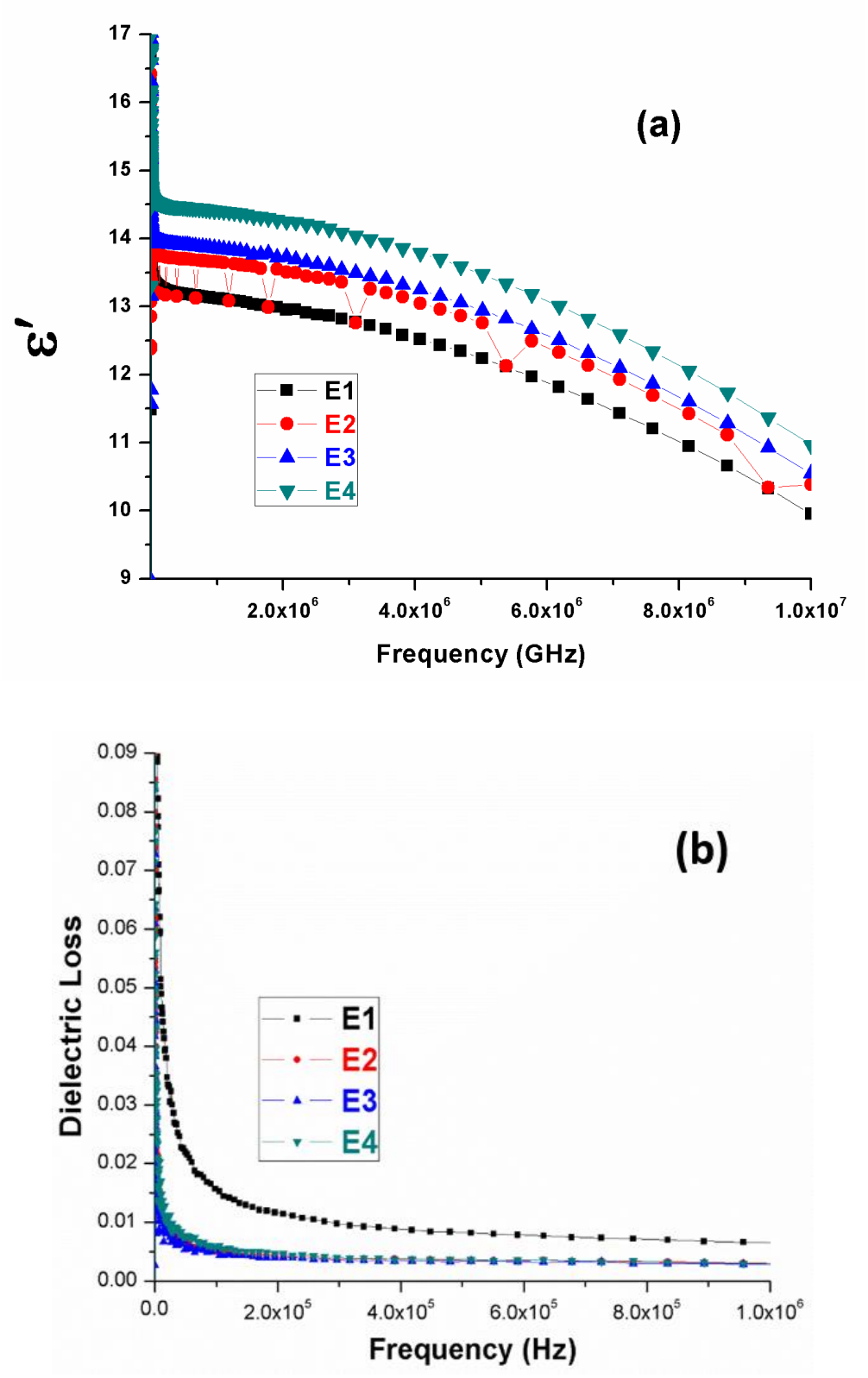
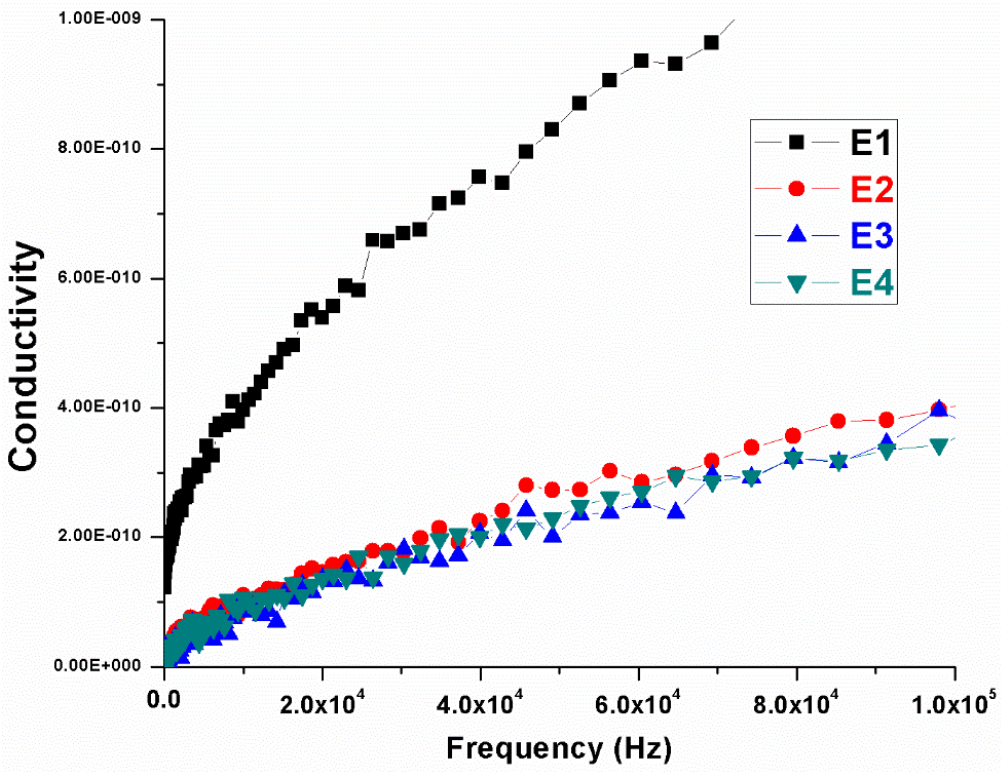


Figure 6. 8: Graph between dielectric constant and loss with frequency (1Hz to 1 MHz).

The decrease in specific resistance or resistivity (ρ) with frequency can be explained by Koop's theorem, which supposed that the dense ceramic material acts as a multilayer capacitor.

The effect of the multilayer capacitor increases with frequency; as a result, the value of ρ decreases. The resistivity increased with BT addition due to the reduction in porosity. The presence of pores in the porcelain composition were directly impact or affect the densification of the samples resulting in the decline of efficient loading area and dielectric property.



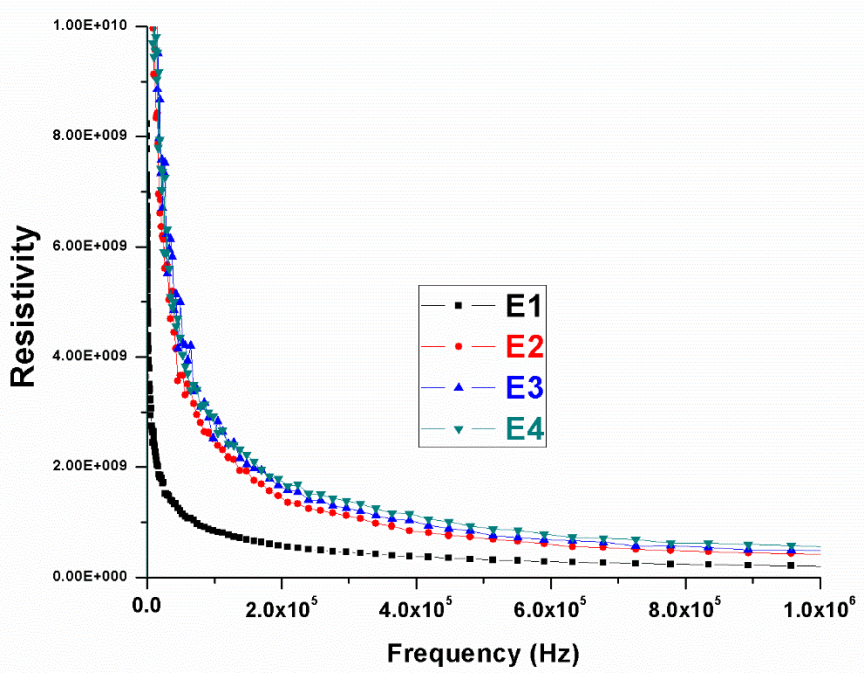


Figure 6. 9: Resistivity or specific resistance and conductivity versus frequency (in Hz) for all the sample sintered at 1350°C.

6.3.4.1 AC Dielectric Permittivity and Dielectric Loss Measurement at Microwave Frequency

It is noted from Figure 6. 10, the AC dielectric permittivity (ε′) values lies from 4.92 to 4.45 for sample E4 and 4.62 to 4.12 for sample E1 in microwave frequency (3 to 20 GHz) at room temperature. The ε′ observed to decreases within the microwave frequencies ranges (3–20 GHz). The ε′ value increases with increasing BT content up to 2 % addition in the base composition. The dependence of dielectric loss or tangent (tanδ) on the frequency is shown in Figure 6. 11. The dielectric loss was found within the range of 0.109 to 0.355 for sample E4, Figure 6. 11.

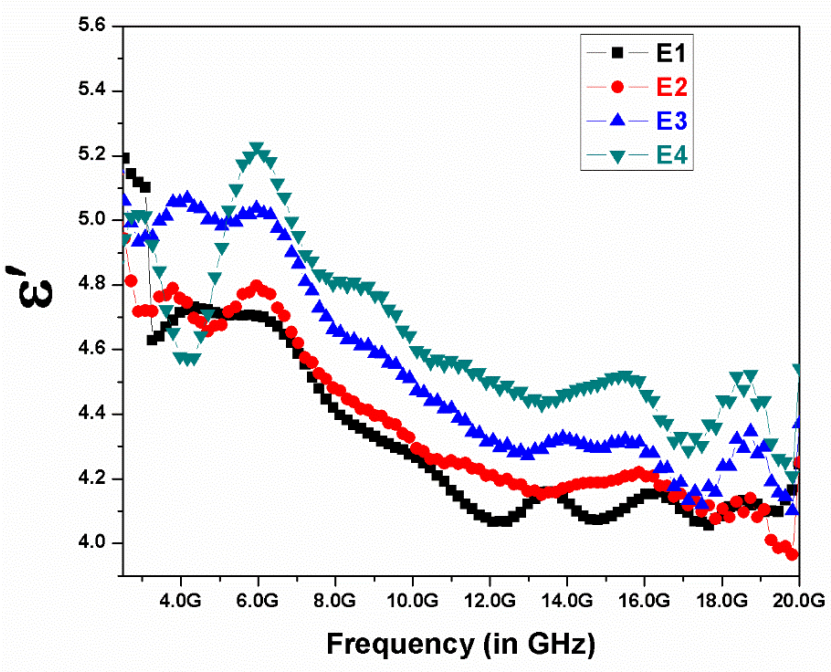


Figure 6. 10: AC Dielectric permittivity versus frequency (GHz) variation of sintered samples investigated at room temperature.

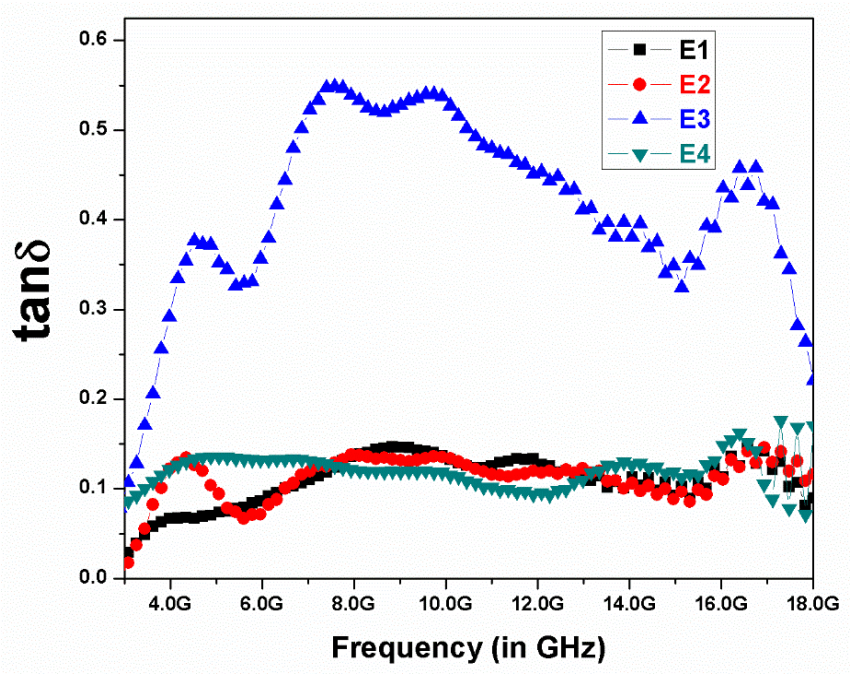


Figure 6. 11: AC dielectric loss (loss tangent) versus frequency (GHz) variation of sintered samples investigated at room temperature.

6.3.4.2 AC Dielectric Strength

The average value of five samples of each composition for AC dielectric strength data is shown in Table 6. 4. Figure 6. 12, reveals that the AC dielectric strength of prepared porcelain insulator slightly increases with increase in BT content. It is observed from Figure 6. 12 with increases concentration of BT (from 0 to 2 wt. %) the ac dielectric strength are constantly increases in slight amount. The relative permittivity of BT is much higher than that of zirconia (≈ 27) and alumina (≈ 10), means having high insulation property. The highest observed AC dielectric strength for samples E4 is 25.39 ± 0.5 KV/mm, which is sintered at 1350°C. It may be concluded that $BaTiO_3$ as a filler is effective in improving electrical properties.

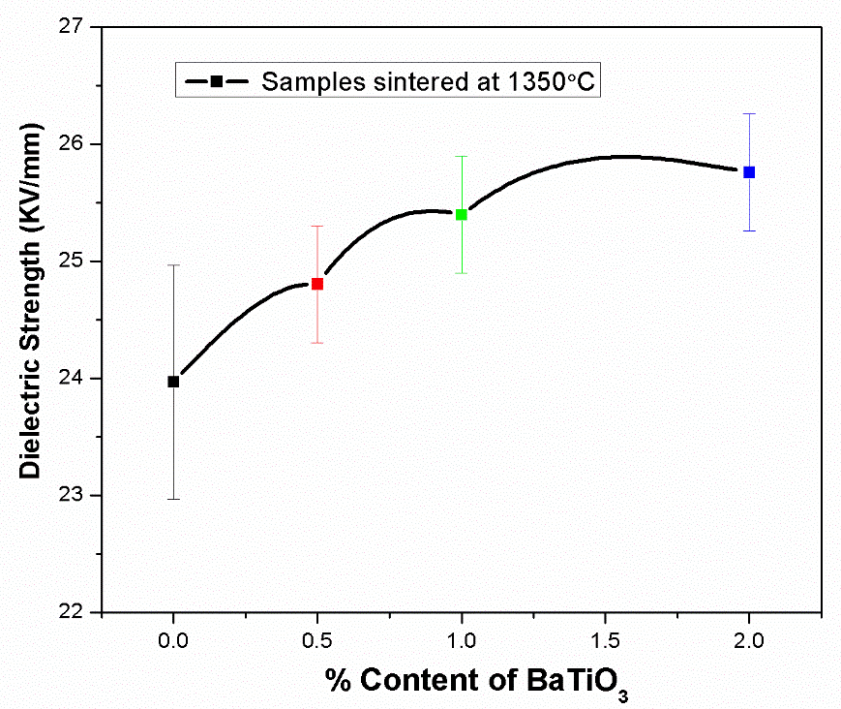


Figure 6. 12: Show the graph for dielectric strength with $BaTiO_3$ concentration.

Table 6. 4: Measurement of dielectric strength of the different samples composition.

Sample	Average Breakdown Voltage of five samples (KV)	Average thickness of five samples (mm)	Dielectric Strength (KV/mm)
E1	29.00	1.21	23.96 ± 1
E2	31.50	1.27	24.80 ± 0.5
E3	32.00	1.26	25.39 ± 0.5
E4	34.00	1.32	25.75 ± 0.5

6.4 Conclusion

The $BaTiO_3$ (BT) addition are responsible for the electrical properties improvement in the prepared ceramic porcelain insulator.

The bulk density increased and porosity decreased when BT content was increased up to 0.5 Wt. % due to the intense bond formation with zirconia, silica, and alumina concentration. The following things were observed when zirconia was replaced with BT in the base porcelain composition by 0.5 wt. %.

- It has been found that the maximum bulk density, the bending, tensile and compressive strength values of porcelains sintered at 1350°C for 2 hours were 2.65 g/cm^3, 144 ± 5 MPa, 40.50 ± 3 MPa and 223 ± 5MPa respectively
- Maximum dielectric strength value observed for 0.5 % BT addition is 24.80 ± 0.5 KV/mm.

After further addition of BT (on 1 and 2 wt. %) on base composition found that mechanical properties of insulator decreases but electrical properties i.e., AC relative

permittivity at low and microwave frequency, and AC dielectric strength of insulator increases. High dielectric constant provides the advantage in power transmission and low dielectric constant used in high-speed electronic circuits (capacitor dielectrics).