

Figure (4-b): Dielectric losses of $BaTi_{0.95}Zr_{0.05}Y_{0.004}O_3$ sintered at $1350^{\circ}C$

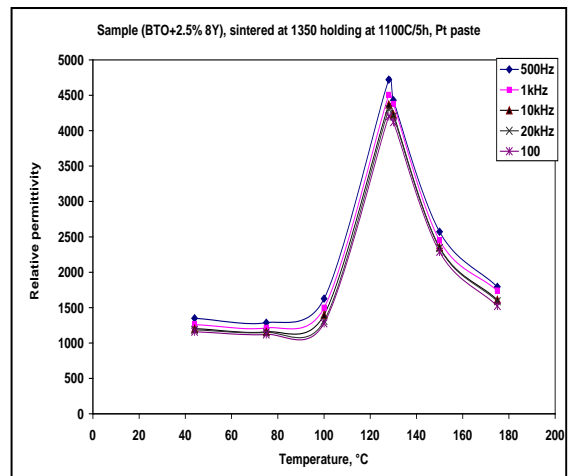
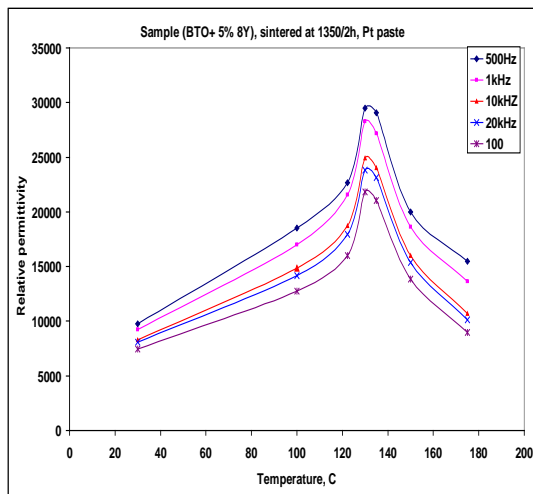
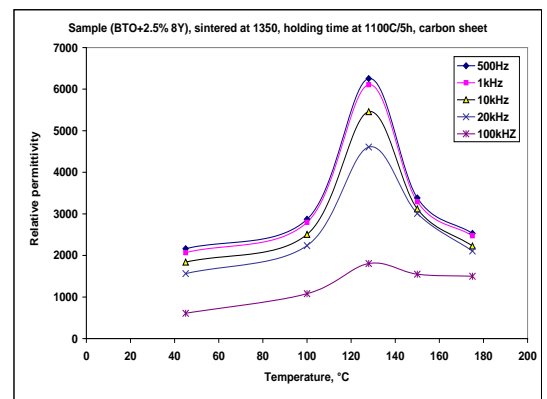


Figure (4-a): Relative permittivity of $BaTi_{0.95}Zr_{0.05}Y_{0.004}O_{3-\delta}$ sintered at $1350^{\circ}C$



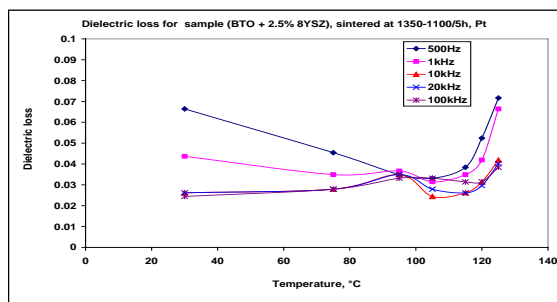


Figure (5): Relative permittivity and dielectric losses of $\text{BaTi}_{0.975}\text{Zr}_{0.025}\text{Y}_{0.002}\text{O}_3$ sintered at 1350°C

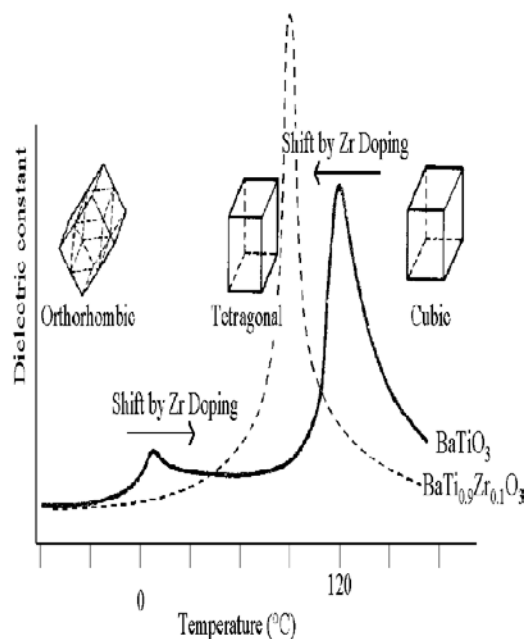


Figure (6): Schematic diagram showing the effect of Zr doping of BaTiO_3 on the dielectric properties⁽⁷⁾

F. Moura et al.⁽⁴⁾ reported that Zirconium is an effective substituent in BaTiO_3 to decrease and shift the Curie temperature. Moreover, Zr^{4+} ion is chemically more stable than Ti^{4+} ion and has a large ionic size to expand the perovskite lattice. Therefore, the substitution of Ti by Zr would depress the conduction by electronic hopping between Ti^{4+} and Ti^{3+} , and it would also decrease the leakage current of the BaTiO_3 system. It was reported that an increase in the Zr content induces a reduction in the average grain size, decrease the dielectric permittivity ϵ_r maintaining a low and stable leakage current^(4,5).

V. Vinothini et al.⁽⁹⁾ reported that BaTiO_3 at room temperature has a Perovskite tetragonal crystal structure with large barium ion (Ba^{2+}) on the corners of the unit cell, oxygen ions (O^{2-}) in the face centers forming an octahedral arrangement and an off-centered small titanium cation (Ti^{4+}). Partial substitution of Ti^{4+} with Zr^{4+} , which has a larger ionic radius, forms the solid solution $\text{BaZr}_x\text{Ti}_{1-x}\text{O}_3$, with an associated increase in the lattice parameter and consequent change in the crystal structure, phase transition temperatures and the dielectric properties compared to pure BaTiO_3 . Previous research performed using submicron-sized powders had suggested that BZT remains tetragonal at Zr additions up to 0.2 mol. %, then change to pseudo monoclinic over the range 0.2 – 0.6 mol. % before

becoming rhombohedral at Zr substitution levels up to ~ 20 mol. %. Above 20 mol.% of Zr additions, BZT is cubic at room temperature⁽⁹⁾. While pure BT undergoes a sharp ferroelectric to paraelectric (tetragonal to cubic) transition at 130°C , BZT ceramics with up to 10 mol.% of Zr exhibit a similar behavior with a shift in the phase transition temperature from 130°C to 110°C ⁽⁷⁾, that is in agreement with our results. V.Vinothini et al.⁽⁹⁾ also added that these phase transition temperatures and phase boundary compositions will be dependent on the crystallite size of the primary particles. As the particle size is reduced to approximately that of the domain size of around tens of nanometers, the particles tend to stabilize to the nearest high symmetry crystal structure (cubic), presumably because of the high surface energy. The critical particle size at which this occurs is still unknown and seems to vary depending on the synthesis route used⁽⁹⁾. V.Vinothini et al.⁽⁹⁾ used a microwave assisted heating to synthesis a nanopowders of BZT with a former requiring lower temperature and shorter times compared to the conventional heating ($700^\circ\text{C} / 30 \text{ min}$, versus $900^\circ\text{C} / 5 \text{ h}$ for the conventional), and added that the microwave synthesized BZT compositions were found to have well crystallized, finer nanoparticles with less agglomeration and higher dielectric permittivity compared to the conventionally prepared powders.

N. Binhayeeniyi et al.⁽¹⁰⁾ prepared BZT, $\text{BaZr}_x\text{Ti}_{1-x}\text{O}_3$ ($x = 0.0 - 0.2$), using a sol-gel process and sintering aid – 1.5 wt% Lithium oxide (Li_2O) and a firing temperature lower by about 30 % to $900^\circ\text{C} / 4 \text{ hrs}$, without any degradation of the electromechanical properties. They⁽⁸⁾ observed a highest dielectric constant beak for $x = 0.05$ at temperature 103°C , about 9500. The curie temperature of the beaks decreased with increasing zirconium values, x , and reached about 40°C at $x = 0.2$.

3. SEM Analysis

SEM micrographs in the back scattered electron mode show the microstructure of the sintered samples, are shown on Figure (7). These micrographs show a relatively dense and homogeneous material. The grain growth of the samples sintered at 1350°C , with holding time for 5 hrs at 1100°C , is higher than that for the samples sintered at 1350°C for 2 hrs.

P.Zheng et al.⁽¹¹⁾ studied the grain-size effects on the dielectric and piezoelectric properties of poled BaTiO_3 ceramics and concluded that dielectric and piezoelectric constant increases significantly at room temperature with the reduction of the average grain size. This can explain the high permittivity constant of the sample sintered at 1350°C for 2 hrs measured at room temperature, Figure (3-a).

4. Conclusions

- (1) The barium zirconate titanate (BZT) ceramics prepared through a solid state reaction between barium titanate and 8Y-cubic stabilized zirconia, sintered at 1300°C were investigated.

- (2) The room temperature X- ray diffraction patterns of the sintered samples show the presence of a single phase structure. The dielectric properties of the different samples had been elucidated by RCL meter as a function of temperature.
- (3) The dielectric constants measured for the different samples showed clear increase of the dielectric constant with some decrease of the curie temperature.
- (4) The microstructure observations by scanning electron microscopy (SEM) revealed that the grain growth depend on the heating schedule used for the sintering.

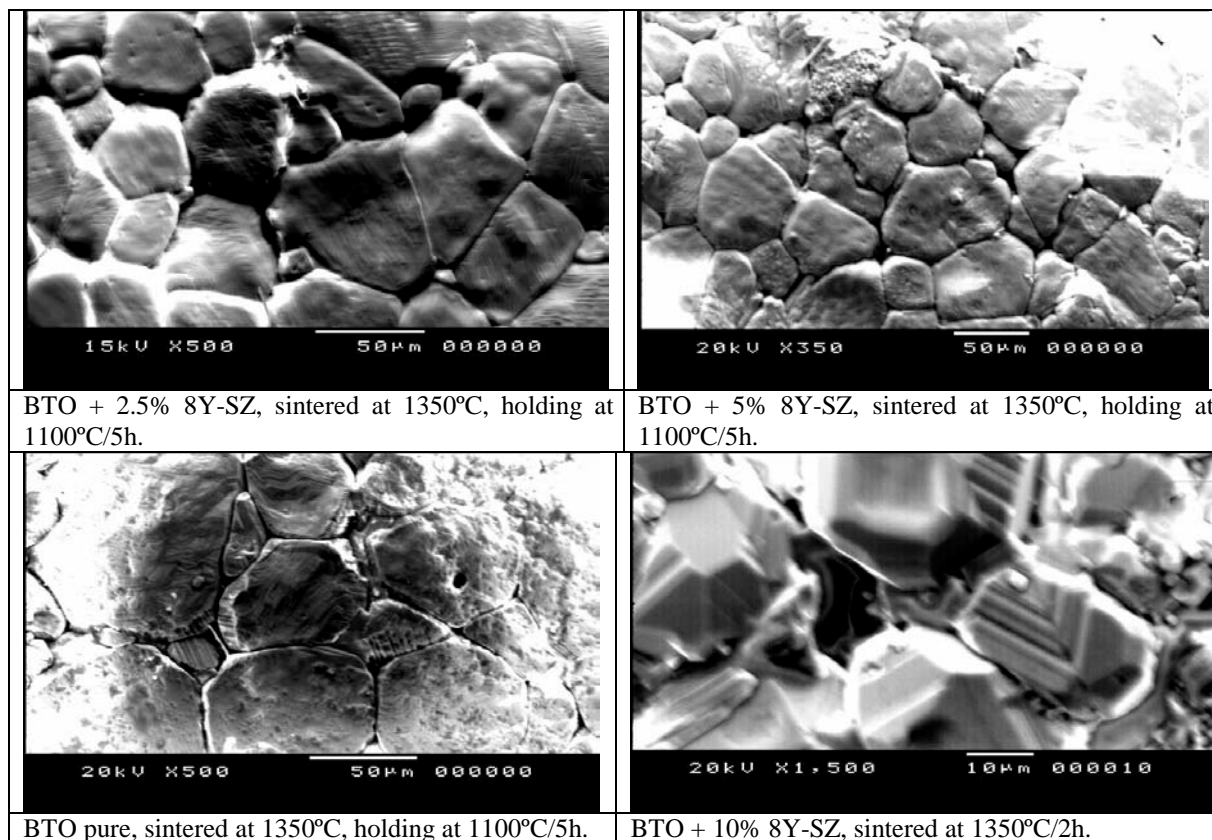


Figure 7: Microstructure by SEM of the different sintered samples

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