



STRUCTURAL, AC CONDUCTIVITY AND DIELECTRIC PROPERTIES OF $Ba_{1-x}Sr_xTiO_3$ CERAMIC COMPOUND

Muthafar F. Al-Hilli

Department of Physics, College of Science, University of Baghdad, Baghdad, Iraq

ABSTRACT

Strontium doped $BaTiO_3$ ceramics with the chemical formula $Ba_{1-x}Sr_xTiO_3$ where $x = 0.01, 0.02, 0.03$ and 0.04 were prepared by conventional solid solution technique and sintered at $1100\text{ }^\circ\text{C}$ for 2 hours. The x-ray diffraction patterns confirmed the formation of a pure perovskite phase. The crystallite size was estimated by Scherrer's equation and found to decrease with strontium addition. The microstructure was studied by scanning electron microscopy. AC conductivity increased with frequency and decreased with substitution ratios of strontium ions. The dielectric constant was noticed to decrease with frequency and increase with strontium addition increase. The dielectric loss factor and dielectric loss tangent decreased with frequency and increased with strontium addition increase.

KEYWORDS: Piezoelectric Ceramics, dielectrics, perovskite.

INTRODUCTION

The superior dielectric constant of $BaTiO_3$ associated with its simple crystalline structure has attracted a great concern in variety of applications e.g. memory cell capacitors in future ultra-large-scale-integrated dynamic random access memories (ULSI-DRAM). Barium titanate doped with Sr is a promising piezoelectric ceramic because of its good insulation properties with high dielectric permittivity as well as low dielectric loss factor close to room temperature^[1,2]. Perovskite-based $Ba_{1-x}Sr_xTiO_3$ alloys are prototype ferroelectrics used extensively in the capacitor industry. The solid solution group of Barium strontium titanate of the chemical formula ($Ba_{1-x}Sr_xTiO_3$) is practically composed of barium titanate and strontium titanate and the Curie temperature of this material covers a broad extent of temperature. The ceramic phase of $SrTiO_3$ in its bulk shape transfers near 105 K from the structure of simple cubic perovskite to the structure of tetragonal perovskite and keeps that phase-structure going down to absolute zero temperature. The phase of $BaTiO_3$ ceramic is a typical ferroelectric material with its Curie temperature around $130\text{ }^\circ\text{C}$ ^[3,4]. However, barium and strontium titanate piezoelectric ceramic demonstrates isomorphism and creates its solid solution covering the whole extent of composition, and this what finally led to the evolution of the ceramic phase of barium strontium titanate (BST). Then, by tailoring the composition of these solid solutions and varying the doping type several properties could be controlled e.g. temperature of transformation, diffuseness, and dielectric constant. The nature of dopant may reduce the structure tetragonality in addition to the transition temperature. The transition diffusive nature is basically ascribed to the behavior of inter- and intragranular strain. Furthermore this ceramic is known by its inhomogeneity of composition which may results in widening of transition. The addition of strontium lowers both of the Curie (T_C) transition and the orthorhombic to tetragonal (T_{o-t})

transformation temperature, while it does not influence other transitions like the rhombohedral to orthorhombic (T_{r-o}). This behavior is basically accounted for the influence of cation size; the smaller Sr leads the cubic phase to be stabilized at lower temperature^[5]. This work reports a study of Sr doping effects on the structure, AC conductivity and dielectric properties of $Ba_{1-x}Sr_xTiO_3$. The possible effects of the composition-dependent properties behavior are discussed.

EXPERIMENTAL

Samples of barium strontium titanate ($Ba_{1-x}Sr_xTiO_3$) where $x = 0.01, 0.02, 0.03$ and 0.04 were prepared by using solid state reaction technique. AR grade powder of $BaCO_3$, TiO and Sr_2O_3 were mixed in the proper molar ratios. The samples with dimensions of (radius= 15 mm and thickness = 5 mm) were prepared by pressing at 3 tons. The pellets were sintered at $1100\text{ }^\circ\text{C}$ for 2 h and cooled to ambient temperature. The dielectric measurements, namely, dielectric constant, Dielectric loss factor and loss tangent were investigated over a range of frequency from 10 kHz to 10 MHz by using an HP 4275A MULTI-FREQUENCY LCR meter. Samples were examined by X-ray diffraction using device miniflex Cu K radiation ($\lambda = 1.540598\text{ \AA}$), at scan speed= 5.0 deg/min. The microstructure was investigated by scanning electron microscope using SEM Hitachi S3400.

RESULTS & DISCUSSION

Figure 1 illustrates the x-ray diffraction patterns of ($Ba_{1-x}Sr_xTiO_3$) where $x = 0.01, 0.02, 0.03$ and 0.04 . It confirmed the formation of a single perovskite phase. It was noticed that with increase in strontium concentration the peaks position shifts towards higher diffraction angles leading to a decrease in the crystallite size as shown in table 1. The crystallite sizes were calculated experimentally by Scherrer's equation with the data obtained from the XRD spectrum. The compositions of the samples phase were not

altered by the increase of Sr content. This may indicate that the increase in strontium substitution did not create any undesirable secondary phases in the structure. However, the relative intensity of the peaks showed a significant improvement by the addition of the Sr ions. Basically the existence of Sr ions executed a positive role

in sintering and crystallization processes of the microstructure [6].

Figure 2 shows the microstructure of the sample with $x=0.03$. The topography shows a homogenous surface structure with no clear imperfection. However, a small amount of porosity may be seen. This is may be due to the low sintering temperature.

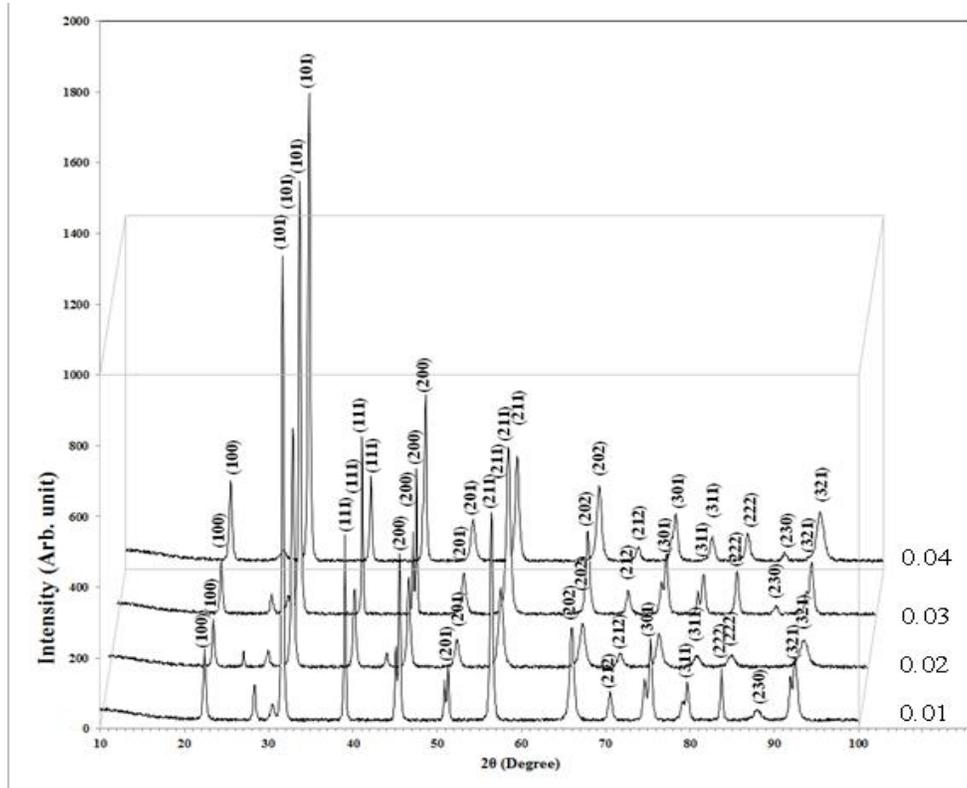


FIGURE 1. x-ray patterns of samples $Ba_{1-x}Sr_xTiO_3$ where $x = 0.01, 0.02, 0.03$ and 0.04 .

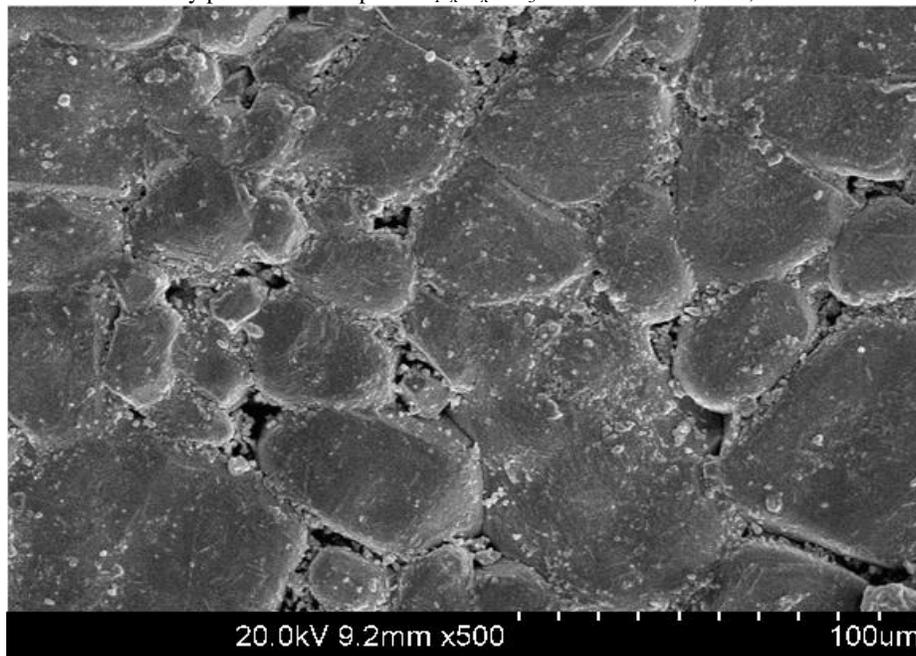


FIGURE 2. SEM microphotograph of the sample $Ba_{0.97}Sr_{0.03}TiO_3$

TABLE 1. Crystallite size of samples $Ba_{1-x}Sr_xTiO_3$ where $x = 0.01, 0.02, 0.03$ and 0.04 .

x	crystallite size ()
0.01	300
0.02	210
0.03	263
0.04	263

Figure 3 shows the change in AC conductivity increases with frequency for different substitution ratios of Sr ions. At low frequencies the AC conductivity was notice to change linearly with frequency. This may be attributed to the DC-like behavior of conductivity at lower frequencies. When frequency was further raised the conductivity was found to increase more rapidly^[7]. The AC conductivity

was noticed to drop with Sr content increase. This is due to the positive temperature coefficient of resistivity (PTCR) influence in $BaTiO_3$ phase which is considered as a phenomenon controlled by the grain boundary of the structure. In particular, Sr-substituted $BaTiO_3$ was found to show the PTCR effect close to room temperature^[8].

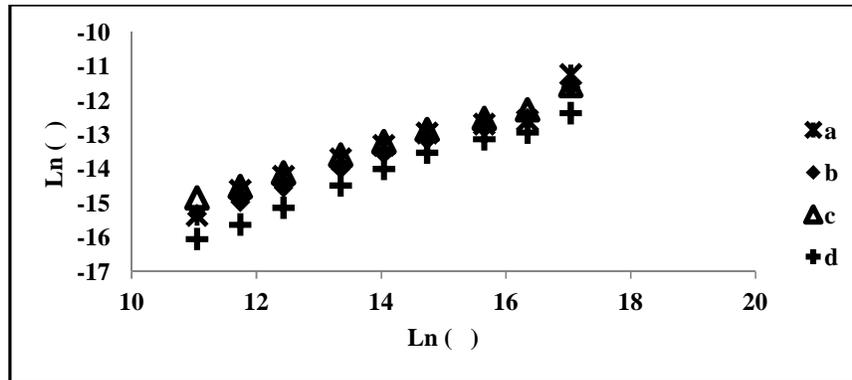


FIGURE 3 shows $\ln \sigma$ versus frequency, where a, b, c and d represents the values of $x=0.01, 0.02, 0.03$ and 0.04 , respectively .

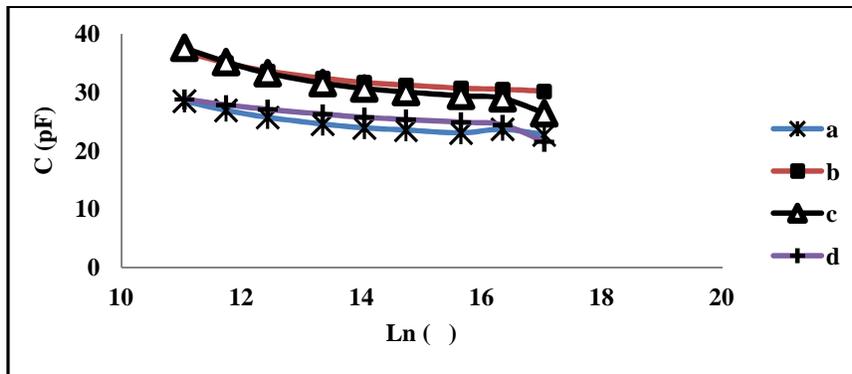


Figure 4 illustrates the change in capacitance with frequency at different Sr content. The capacitance was noticed to decrease with frequency and increases with Sr content increase.

Fig.4 shows the Capacitance (PF) versus frequency, where a, b, c and d represents the values of $x=0.01, 0.02, 0.03$ and 0.04 , respectively.

Figure 5 shows the dielectric constant relation with frequency at different Sr content. The dielectric constant was noticed to fall down with frequency and increases with strontium addition increase. This may be explained on the model of internal boundary layer capacitance. This model proposes an inverse proportionality of dielectric constant with the ratio of grain boundary to the crystallite size. A larger grains existence allows a greater number of crystal imperfections to exist more than in smaller grains. So the change in dielectric constant is correlated to the change in crystallite size (see table 1)^[9].

Figure 6 shows the dependence of dielectric loss factor upon frequency at different Sr content. The dielectric loss factor was noticed to decrease with frequency and

increases with Sr content increase. Basically, the electronic exchange between different ions of Ti^{4+} and Ti^{3+} ions produces an n-type charge carriers, however, the charge drift mobility of p-type carriers is usually lower than that charge carriers of n-type. It was noticed that at higher frequencies the dielectric loss factor decreased due to the decrease in dipole contributes to the polarization [10]. Figure 7 shows the relationship between dielectric loss tangent and frequency at different Sr content. It is evident that the behavior of dielectric loss tangent is similar to that of dielectric loss factor for the different addition ratios of strontium ions.

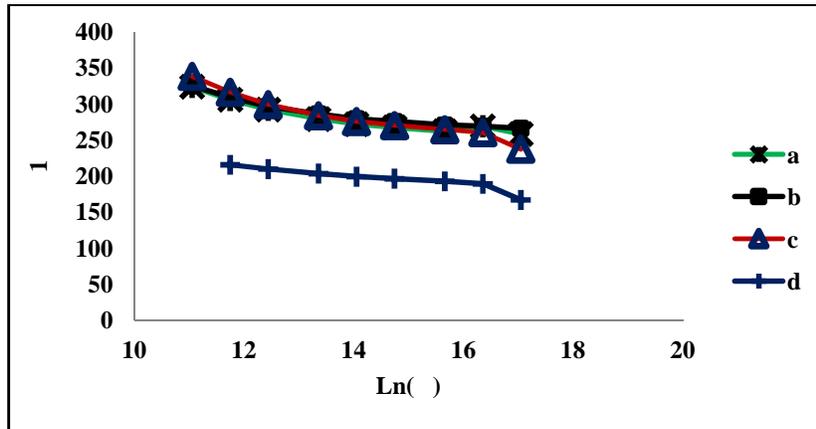


FIGURE 5: show dielectric constant versus frequency, where a, b, c and d represents the values of $x=0.01, 0.02, 0.03$ and 0.04 , respectively.

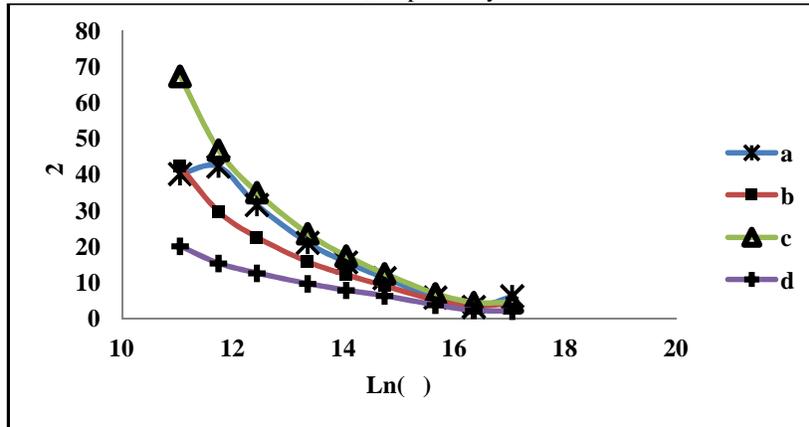


FIGURE 6: show dielectric loss versus frequency, where a, b, c and d represents the values of $x=0.01, 0.02, 0.03$ and 0.04 , respectively.

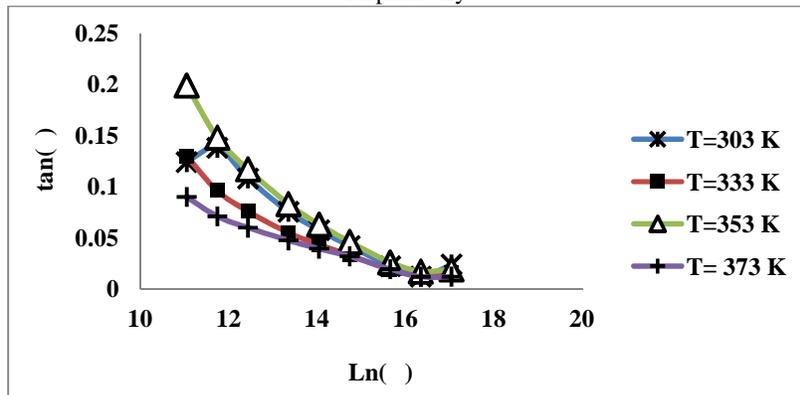


FIGURE 7: shows dielectric loss tangent versus frequency at different temperatures.

CONCLUSION

The crystallinity of $Ba_{1-x}Sr_xTiO_3$ increases with increase in strontium content. AC conductivity measurement confirms the transition of Curie temperature near room temperature suggesting that this material is very promising candidate for gas and humidity sensors application. The dielectric behavior of the material showed a remarkable dependence upon frequency and composition. The reduced and nearly constant dielectric loss near room temperature makes this material suitable for variety of applications.

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