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Characterization of $Ba_{0.35}Sr_{0.65}TiO_3$ Made by Sol-Gel Method with X-Ray Powder Diffraction, Field Emission Scanning Electron Microscopy and Impedance Spectroscopy in Capacitor Application

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Characterization of $Ba_{0.35}Sr_{0.65}TiO_3$ Made by Sol-Gel Method with X-Ray Powder Diffraction, Field Emission Scanning Electron Microscopy and Impedance Spectroscopy in Capacitor Application

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Abstract. A thin film of Barium Strontium Titanate (BST) of ferroelectric material of $Ba_{0.35}Sr_{0.65}TiO_3$ has been made using sol-gel method and annealed at 600, 650, and 700°C in order to obtain its crystalline structure. This thin film was characterized using X-Ray Diffraction (XRD), Field Emission Scanning Electron Microscopy (FESEM) and Impedance Spectroscopy. The results of XRD characterization show a graph of intensity against angle 2θ. The XRD patterns of $Ba_{0.35}Sr_{0.65}TiO_3$ have many peaks and confirmed that the material of $Ba_{0.35}Sr_{0.65}TiO_3$ has cubic crystalline structure and their lattice parameter is in average of 2.399Å. The results of FESEM characterization show that the thickness of thin films of $Ba_{0.35}Sr_{0.65}TiO_3$ at 600, 650, and 700°C were 21.21 nm, 34.61 nm and 39.08 nm, respectively. The values of capacitance at the frequency of 100 Hz with temperature of 600, 650, and 700°C are 4.84×10^{-11}F, 5.24×10^{-11}F and 5.61×10^{-11}F, respectively. The dielectric constant of the thin films of BST at the frequency of 100 Hz with temperature of 600, 650, and 700°C are 5.47, 5.90 and 6.28, respectively. While the loss of dielectric values of the thin films of BST at 100 Hz with temperature of 600, 650, and 700°C are 0.03, 0.08 and 0.12, respectively. The result of characterization by Impedance Spectroscopy shows that the higher the frequency, the smaller complex capacitance, dielectric constant and dielectric loss. Generally, the higher annealed temperature, the higher complex capacitance and dielectric constant.

1. Introduction

Along with the rapid development of the industry, the needs for electronic devices are getting smaller with increasingly large and advance performances. Thin layer has an important role in its development because it is widely used in applications such as sensors for energy storage, detectors, capacitors and thin films. One of the ferroelectric thin film layers is Barium Strontium Titanate (BST) [1,2].

The ferroelectric material has an ability to change the direction of its internal electricity. The ferroelectric material, especially the mixture of Barium Strontium Titanate (BST), has high hysteresis properties and dielectric constants. It can be applied to sensor and memory actuators. The ferroelectric material, which has the most interesting mixture of properties for memory applications, is Barium Strontium Titanate (BST). This material has many advantages such as having high dielectric constant, and low dielectric loss. The high dielectric constant will increase the higher charge capacitance [3].

The capacitor is one of the electronic components that can store the charge or electrical energy. The capacitor is divided into several types; one of them is parallel plate capacitor. One of the researches that has been conducted is dealing with a parallel plate capacitor. It has a capacitance value that can be made larger if the distance between the plates is reduced, the area of the plate is enlarged, and the dielectric material Barium Strontium Titanate (BST) can be further enlarged.

2. Materials and Method

The manufacture of BST thin film was done from a mixture of barium carbonate, strontium carbonate and titanium(IV) Isopropoxide. The BST was prepared by using sol-gel method then continued by using spin coating and annealing process at 600°C, 650°C and 700°C. The thin films that have been produced were characterized using XRD (Cu λ = 1.541Å), FESEM and impedance spectroscopy.

The procedure was started with the preparation of 30 pieces of glass substrate of 1 × 2 cm. The substrate that has been cut then washed using aqua DM and ethanol 96% solution and soaking in ultrasonic bath. Substrate glass that has been soaked then dried in oven for 15 minutes. The construction of BST thin films shown in Figure 1, then applied to the capacitor with an electrode. The electrodes used was aluminium (Al), in which Al was dipped in acetyl acid. Next, the glass and Al electrode were put together, the left side of the glass was covered with aluminium foil, and the middle part will be dripped with BST solution. After the annealing process, the BST and Al portions are on the left and covered with aluminium foil while dripped Al solution was on the right of the glass substrate. The BST thin film then applied to the capacitor.
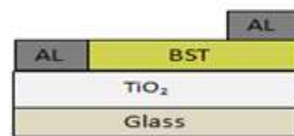

Figure 1. The construction of $Ba_{0.35}Sr_{0.65}TiO_3$.

3. Result and Discussion

The diffraction pattern of the thin film layer of $Ba_{0.35}Sr_{0.65}TiO_3$, presented in Figure 2, shows that it has a cubic plane and several peaks due to its high crystalline structure. The emerging peaks indicate a certain field orientation that can be calculated. These peaks indicate a meaningful crystal having an orderly arrangement of particles. Differences annealing temperatures gives different intensity of diffraction in which the intensity is getting higher as the temperature increases but still have the same Miller Indices (h, k, l) and diffraction angle (2θ). Table 1 indicates that the distance between planes (d_{hkl}) decreases as the diffraction angle (2θ) increases.

Figure 2a, 2b, and 2c show relationship between intensity and diffraction angle (2θ). The higher annealing temperature only affect the intensity. High intensity indicates higher crystalline level, since at high temperatures, the atoms are easier to diffuse with other atoms. As a result, the bonds between atoms will be stronger and more regular [4-7].

Table 1. Powder XRD data.

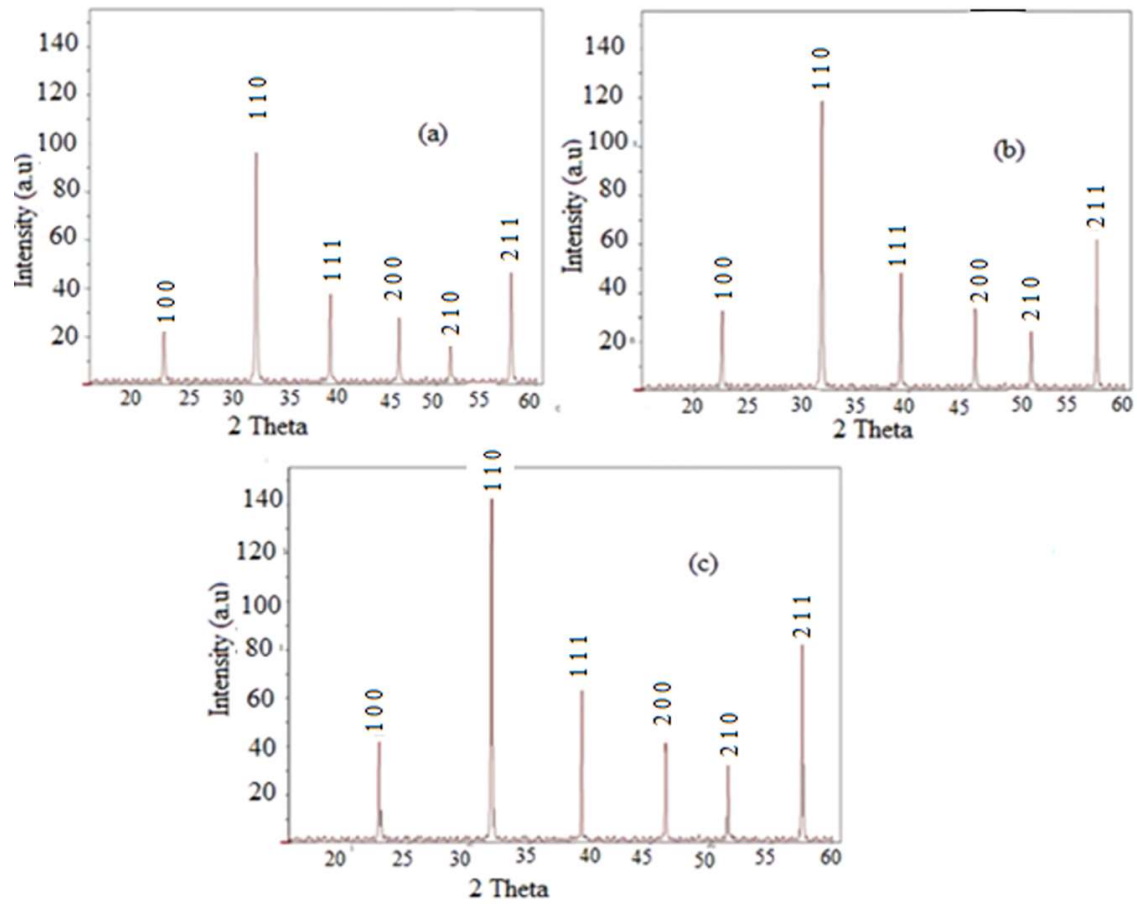| No	(2θ) (°)	θ (°)	d_{hkl}(Å)	Lattice unit (Å)
1	22.54	11.27	3.95	3.952
2	31.84	15.92	2.81	3.977
3	39.30	19.65	2.29	3.971
4	46.23	23.11	1.96	3.922
5	51.44	25.72	1.77	3.971
6	57.54	28.77	1.60	3.924

Figure 2. The relationship between peak intensity and diffraction angle (2θ) at annealing temperature of (a) 600°C, (b) 650°C and (c) 700°C.
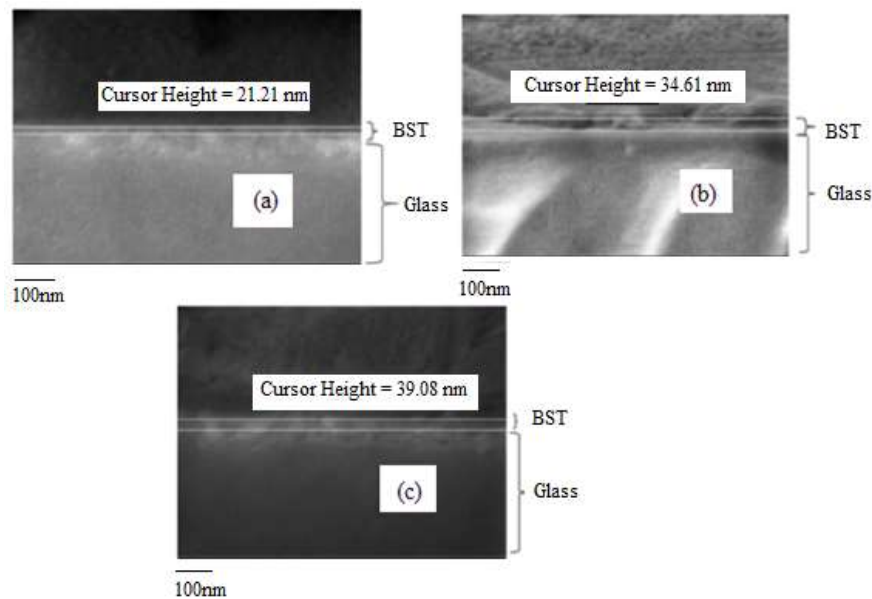

Figure 3. Thickness of BST layer of the capacitor $Ba_{0,35}Sr_{0,65}TiO_3$ on the glass substrate at annealed temperature of (a) 600°C, (b) 650°C and (c) 700°C.

Figures 3a, 3b, and 3c show the thickness of BST thin films on the substrate surface which are 21.21, 34.61, and 39.08 nm, respectively. Figures 3 indicate that the thickness of BST thin film is increasing as the annealing temperature increased, which is due to the growing size of the grains at high temperatures. Kingery *et al*. (1991) state that the increase of annealing temperature can increase the movement of atoms, causing small grains to combine to form larger grains [8,9]. A DLC (Diamond-like carbon) of thin film that is deposited using DC-PECVD, meanwhile, the thickness of the substrate is directly proportional to the annealing temperature. The higher annealing temperature provided causes the increase of thickness of DLC thin film glass substrate [10].

The relationship between complex capacitance, dielectric constant, and dielectric loss with frequency in various annealing temperatures (600, 650, and 700°C) are presented in Figure 4. The complex capacitance at the frequency of 100 Hz with temperature of 600, 650, and 700°C are 4.84×10^{-11} F, 5.24×10^{-11} F and 5.61×10^{-11} F, respectively. The dielectric constant of the thin films of BST at the frequency of 100 Hz with temperature of 600, 650, and 700°C are 5.47, 5.90, and 6.28, respectively. Meanwhile the dielectric loss values of the thin films of BST at 100 Hz with temperature of 600, 650, and 700°C are 0.03, 0.08, and 0.12, respectively. It is also observed that complex capacitances, dielectric constants, and measured dielectric losses decreases significantly as the frequency increased. The higher the frequency, the lower the complex capacitance value, the dielectric constant and the dielectric loss obtained. On the contrary, the lower the frequency, the higher the complex capacitance value, the dielectric constant and the dielectric loss observed. The value of the dielectric constant is directly proportional to the complex capacitance. The obtained dielectric loss value is also lower than the dielectric constant [11]. Dielectric loss at YIG depends on sintering temperature at 1kHz and 1MHz frequencies and increases with the addition of sintering temperature [12]. These conditions are in principle caused by the interface policing process [13].
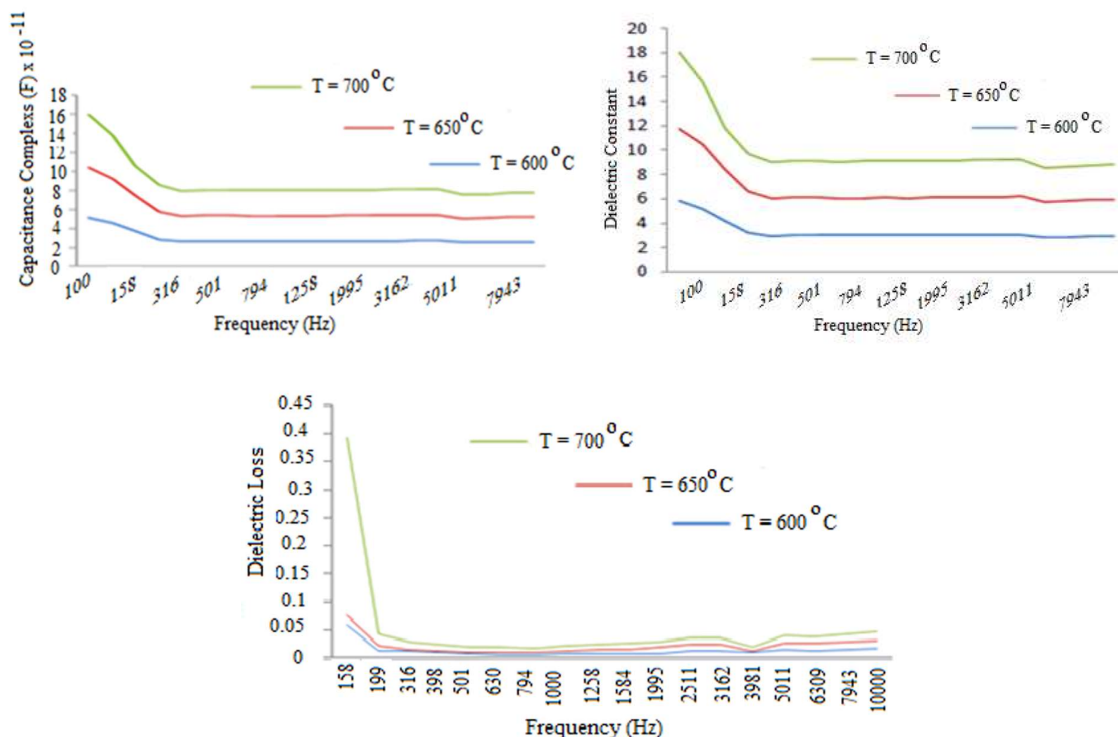

Figure 4. The graph of capacitance complex (top left), dielectric constant (top right), and dielectric loss against frequency of $Ba_{0.35}Sr_{0.65}TiO_3$ capacitor at temperature of 600°C, 650°C and 700°C.

4. Conclusion

The XRD characterization of BST shows that the peak intensity value is getting higher as the temperature increased, but the Miller indices and diffraction angle (2θ) remain the same. Based on the results of FESEM characterization, the BST thin film composition of $Ba_{0.35}Sr_{0.65}TiO_3$ has a thickness of 21.21 nm, 34.61 nm and 39.08 nm, respectively. The result of characterization by Impedance Spectroscopy shows that complex capacitance, dielectric constant and dielectric loss are inversely proportional to a given frequency, in which the higher the frequency, the smaller complex capacitance, dielectric constant and dielectric loss values. The highest values for all three parameters were obtained at the smallest frequency (100Hz).

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