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EFFECT OF SINTERING TEMPERATURE ON THE MICROSTRUCTURE AND MICROWAVE DIELECTRIC PROPERTIES OF STRONTIUM TITANATE ($SrTiO_3$) CERAMICS

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ABSTRACT

In this study, microstructure and microwave dielectric properties of strontium titanate ($SrTiO_3$) ceramics prepared via conventional solid state reaction route were investigated. The starting materials used for the preparation of $SrTiO_3$ were strontium carbonate ($SrCO_3$) and titanium oxide (TiO_2). $SrTiO_3$ ceramics were calcined at 1100 °C and sintered at 1200 °C, 1300 °C, and 1400 °C respectively. Identification of crystalline compounds of $SrTiO_3$ ceramics were analyzed using X-ray Diffraction (XRD). Microstructure of $SrTiO_3$ ceramics were then characterized by Field Emission Scanning Electron Microscope (FE-SEM). The microwave dielectric properties of $SrTiO_3$ were measured in the frequencies range from 1 MHz to 1.5 GHz using Agilent RF Impedance/Material Analyzer 4192B. From the XRD patterns of $SrTiO_3$ ceramics, it showed that $SrTiO_3$ ceramics were in crystalline structure after sintering. $SrTiO_3$ ceramic sintered at 1400 °C has larger grain size with lower porosity. Loose and porous structure was observed for $SrTiO_3$ ceramic sintered at 1200 °C. For $SrTiO_3$ ceramic sintered at 1400 °C, it was observed that the dielectric constant was the highest. Lower dielectric constant was observed for $SrTiO_3$ ceramic sintered at 1300 °C.

Keywords: Ceramics; Dielectric properties; X-ray diffraction; Strontium titanate; Microstructure;

INTRODUCTION

Currently, research on strontium titanate ($SrTiO_3$) are still widely investigated and developed due to its unique properties towards industrial uses. $SrTiO_3$ is a paraelectric material with cubic perovskite-type crystal structure at room temperature [1]. $SrTiO_3$ has many interesting properties and can be potentially used for many applications. $SrTiO_3$ has applications as in dynamic random access memories (DRAM) with 1 Gbit density or higher [2, 3], microelectronics [4], grain boundary layer capacitor (GBLC) [5], photoelectric and dielectric material [6]. $SrTiO_3$ is an attractive material for many high frequency and microwave frequency applications due to its high dielectric constant

that increases on cooling [7]. Furthermore, microwave dielectrics are in demand owing to its low microwave dielectric loss [8].

There are several methods for the synthesis of $SrTiO_3$, which are solid state reaction method, wet chemical method such as sol gel, colloidal or cluster solutions, and gas phase reaction method such as chemical vapor deposition (CVD). In this research, solid state reaction method is used to prepare $SrTiO_3$. This oldest ceramic method has the advantages include low cost of production, reproducible, available precursors, and simplicity of the processes [9].

EXPERIMENTAL METHOD

Preparation of $SrTiO_3$ ceramics

In this research, strontium titanate ($SrTiO_3$) ceramics were prepared by conventional solid state reaction method. The starting powders used to form $SrTiO_3$ compounds were strontium carbonate, $SrCO_3$ (≥99.9% purification), and titanium oxide, TiO_2 (99.6% purification). Initially, the amount of the starting powders of $SrCO_3$ and TiO_2 that were needed to form $SrTiO_3$ compounds was calculated based on the chemistry equation stated in (1) as follows:

$$SrCO_3 + TiO_2 \rightarrow SrTiO_3 + CO_2 \qquad (1)$$

Stoichiometric amounts of powders needed were weighed according to the weight ratio using an electronic balance. The mixture of both powders was then dry mixed and milled for 24 hours in the milling machine in order to achieve the desired particle size distribution. $SrTiO_3$ powders were calcined at 1100 °C for 10 hours using a carbolite furnace.

The purpose of calcination process was to decompose the higher oxides and carbonate in order to reduce the production of gases in the final sintering process. After calcination process was done, $SrTiO_3$ ceramics were crushed and grinded using a mortar for a few hours, followed by the sieving process. In the sieving process, $SrTiO_3$ were sieved using a 45 μm test sieve to ensure homogeneity. $SrTiO_3$ were then pressed into pellets form by the Carver Manual Pellet Press with an applied load of 5.0 metric tons. Finally, $SrTiO_3$ pellets were sintered at 1200 °C, 1300 °C, and 1400 °C respectively using a carbolite furnace.

Characterizations and Dielectric Measurements of $SrTiO_3$ ceramics

The crystalline compounds of $SrTiO_3$ pellets were examined using X-ray Diffraction (XRD) with scattering angles from 20° to 80° in 2θ. The microstructures of $SrTiO_3$ ceramics were observed using Field Emission Scanning Electron Microscope (FE-SEM). For dielectric measurements, the dielectric properties of $SrTiO_3$ ceramics were measured using Agilent RF Impedance/Material Analyzer 4192B in the microwave frequencies range from 1 MHz to 1.5 GHz.

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RESULTS AND DISCUSSION

Figure 1 showed the XRD patterns of $SrTiO_3$ ceramics sintered at different temperatures. From the XRD analysis, it was observed that single phase cubic perovskite structure was formed in all the samples. $SrTiO_3$ has three strongest peaks at 2θ= 32°, 47°, and 57°. $SrTiO_3$ ceramic sintered at 1400 °C has the highest intensity peak as compared with $SrTiO_3$ ceramics which were sintered at 1200 °C and 1300 °C. This series agreed with 01-086-003 of the International Center Diffraction Data (ICDD) standard that confirmed $SrTiO_3$ structures [10]. No secondary phases were found in this XRD analysis indicating the phase formation of $SrTiO_3$ ceramics were completed during the process. $SrTiO_3$ ceramic sintered at 1400 °C showed the highest intensity indicating the highest $SrTiO_3$ content [11]. $SrTiO_3$ ceramic sintered at 1300 °C showed the lowest intensity, thus indicating the lowest $SrTiO_3$ content.

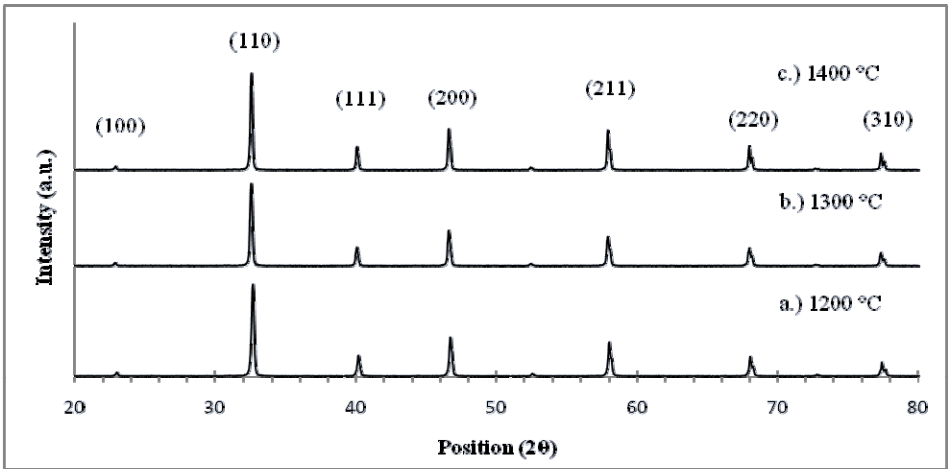

Figure 1: XRD patterns of $SrTiO_3$ ceramics sintered at: (a) 1200 °C, (b) 1300 °C, and (c) 1400 °C

The FE-SEM micrographs of $SrTiO_3$ ceramics sintered at different sintering temperatures were illustrated in Figure 2. Generally, $SrTiO_3$ ceramics sintered at 1200 °C, 1300 °C, and 1400 °C exhibited uniform microstructure and normal grain growth, which all grains grew at roughly the same rate in an uniform manner. FE-SEM analyses performed on $SrTiO_3$ ceramic sintered at 1200 °C showed relatively smaller grain sizes, more grain boundaries, and have more porosity with large number of small grains. As the sintering temperatures were increased, the grains size and density of $SrTiO_3$ ceramic also increased. Thus the concentration of porosity was found to decrease with increasing sintering temperature [12]. $SrTiO_3$ ceramic sintered at 1400 °C exhibited the largest grain size as compared with others. It also revealed total lack of porosity and dense microstructure if compared with others.

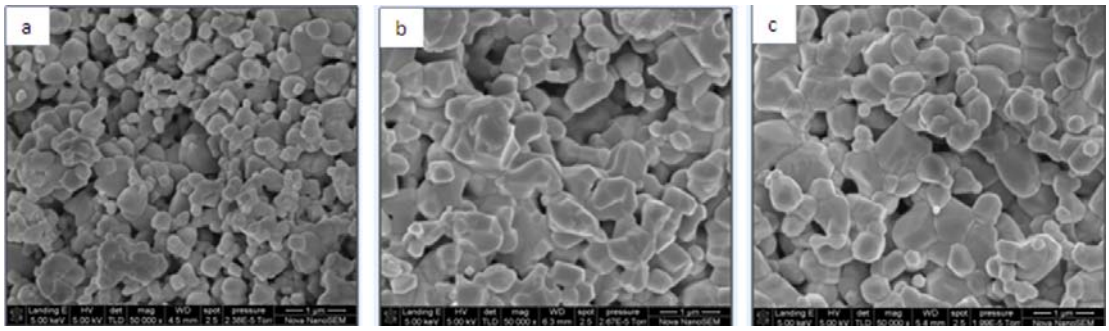

Figure 2: FE-SEM micrographs of $SrTiO_3$ ceramics sintered at: (a) 1200 °C, (b) 1300 °C, and (c) 1400 °C

Table 1 showed the data of dielectric constant and dielectric loss of $SrTiO_3$ ceramics sintered at different temperatures. All of the samples showed almost constant value of dielectric constant along the microwave region. On the other hand, inconsistent value of dielectric loss along the microwave region was observed.

Table 1: Dielectric constant and dielectric loss values of $SrTiO_3$ ceramics sintered at different sintering temperatures

Frequency (Hz)	Sintering Temperature (°C)					
	1200		1300		1400	
	ε_r'	ε_r''	ε_r'	ε_r''	ε_r'	ε_r''
1×10^6	40.33	1.92	25.14	1.17	43.8	2.06
1×10^7	43.61	0.11	26.9	0.05	47.3	0.01
1×10^8	43.35	0.01	26.8	0.00	47.1	0.07
1×10^9	46.01	0.45	26.31	0.12	47	0.74

Figure 3 depicted the variation of dielectric constant with frequency at different sintering temperatures. $SrTiO_3$ ceramics sintered at 1200 °C, 1300 °C, and 1400 °C showed the same trend in microwave frequency region. From the graph, it was observed that all the $SrTiO_3$ ceramics have constant dielectric value in the frequency range 10 MHz to 1 GHz. The increment after 1 GHz could be due to the presence of resonance [13]. $SrTiO_3$ ceramic sintered at 1400 °C has the highest dielectric constant, approximately around 47. This could be due to better grain growth with better diffusion among grains resulting in an increase in the dielectric constant. For $SrTiO_3$ ceramic sintered at 1200 °C, the dielectric constant was reduced owing to higher porosity, which the sample could be filled with air into the pores [14]. $SrTiO_3$ ceramic sintered at 1300 °C exhibited the lowest dielectric constant, around 26. This might be caused by the lack of $SrTiO_3$ phase inside the sample, which might decrease the dielectric constant. This is consistent with XRD results, in which a linear relationship was found to exist between dielectric constant and crystallinity [15].

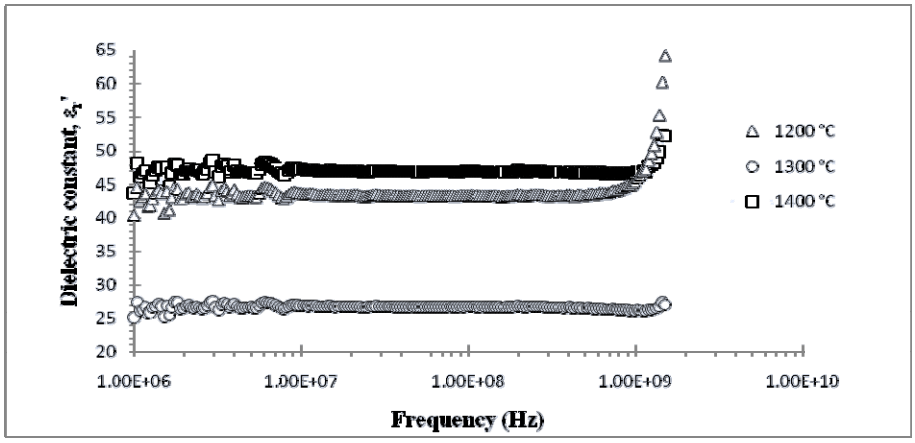

Figure 3: Graph of dielectric constant versus frequency for $SrTiO_3$ ceramics sintered at different sintering temperatures

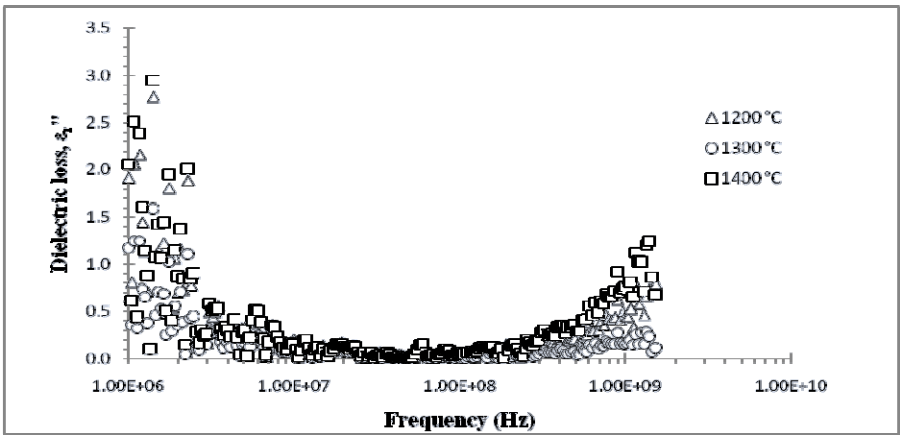

Figure 4: Graph of dielectric loss versus frequency for $SrTiO_3$ ceramics sintered at different sintering temperatures

Figure 4 depicted the variation of dielectric loss with frequency at different sintering temperatures. $SrTiO_3$ ceramics sintered at 1200 °C, 1300 °C, and 1400 °C showed the same trend in microwave frequency region. Along the frequency range from 1 MHz to 10 MHz, the higher value of dielectric loss is observed. As frequency increased, more and more dipoles were unable to follow the direction of oscillating field, thus dielectric loss decreased with increasing frequency [16]. The higher value of dielectric loss at lower frequency could also be due to the presence of space charge polarization between the samples and electrodes (Maxwell-Wagner polarization) [17], which is mostly dominant in this lower frequency range [18]. In the frequency range from approximately 10 MHz to 100 MHz, the values of dielectric loss were in the order 0 to 1.It can be said that low loss $SrTiO_3$ ceramics has been achieved. The dielectric loss for $SrTiO_3$ sintered at 1400 °C was the highest among others. The highest dielectric loss can be attributed to the role of larger grain size. In larger grains, the domain walls can

move easily due to the reduction of grain boundaries [19]. Thus, larger eddy current was induced [19]. Hence, the dielectric loss was increased.

CONCLUSION

$SrTiO_3$ ceramics were successfully prepared by conventional solid state reaction method. XRD analysis of $SrTiO_3$ ceramics revealed the formation of single phase cubic perovskite structure. For microstructural analysis, higher porosity and small grain size were observed for $SrTiO_3$ ceramic sintered 1200 °C. $SrTiO_3$ ceramic sintered at 1400 °C showed larger grain size with lower porosity. For all the $SrTiO_3$ ceramics sintered at 1200 °C, 1300 °C, and 1400 °C, the samples have constant dielectric constant within the microwave frequency region with lower dielectric loss in the microwave frequency range from 10 MHz to 100 MHz. The low loss $SrTiO_3$ ceramic materials in the microwave frequency region can be potentially used as dielectric resonator and substrates. The highest dielectric constant was around 47, which was sintered at 1400 °C. The lowest dielectric constant was around 26, which was sintered at 1300 °C. The dielectric loss for $SrTiO_3$ sintered at 1400 °C showed the highest value, followed by $SrTiO_3$ ceramics sintered at 1200 °C, and 1300 °C.

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