Effect of Sintering Temperatures on the Microstructure and Dielectric Properties of SrTiO$_3$

Leow Chun Yan, Jumian Hassan, Mansor Hashim, Wong Swee Yin, Tan Foo Khoon and Wong Yick Jeng

Department of Physics, Faculty of Science, Universiti Putra Malaysia, Malaysia
Institute of Advanced Technology, Universiti Putra Malaysia, Malaysia

Abstract: Conventional solid state reaction method is a common and effective way to fabricate modern ceramics. For ceramic processing, sintering is an important factor that affects the microstructure evolution, thus optimizing its natural abilities. In this work, Strontium Titanate (ST) ceramic samples sintered at different sintering temperatures were prepared and analyzed by X-Ray Diffraction (XRD) and Scanning Electron Microscope (SEM). XRD is used to determine the samples crystallization, while SEM was for microstructure analysis. Dielectric properties of SrTiO$_3$ samples were measured using Agilent 4291B Impedance/material Analyzer in the frequency range of 1 MHz to 1.5 GHz at room temperature. The dielectric constant is constant with respect to frequency and increased after 1 GHz. Dielectric constant of ST increased with increasing sintering temperatures. ST sintered at 1200°C has the highest dielectric constant of 50. The average grain size increases with increasing sintering temperature.

Key words: Strontium Titanate · Ceramics · Dielectric Properties · Sintering Temperature · Microstructure · Microwave Frequency

INTRODUCTION

Strontium titanate is a non-conducting, quantum paraelectric [1,2] material with cubic perovskite structure. ST based material is suitable for tunable microwave devices due to its high dc-electric-field dependence on the dielectric constant [3]. Also, with good insulating properties [4], this makes it essential in the application of dynamic random access memories [5].

From earlier studies, it has been shown that doped (manganese or antimony) and undoped ST has an influence on the samples dielectric properties [3,6]. ST is a promising material in microwave communication application. ST paraelectric dielectric behavior in the microwave region is an interesting topic to investigate. Generally, dielectric properties of ST ceramic are highly dependent on the formation of its microstructure. In ceramics, sintering is always an important processing parameter that influences ceramics microstructure evolution, grain growth and densification [7,8]. In this study, dielectric properties of the samples sintered at different temperatures (1000°C-1400°C) were investigated with respect to its microstructure.

EXPERIMENTAL: Conventional solid state reaction method was used to fabricate SrTiO$_3$ samples. Highly pure powders of SrCO$_3$ (99.9%) and TiO$_2$ (99.9%) were used as the starting materials. The raw materials were weighed in stoichiometric proportions, dry ball-milled and calcined at 800°C, 1000°C and 1200°C for 10 hours. The calcined powders were ground and sieved to ensure the particle size was homogeneous. The obtained powders were pressed into pellets of diameter 20.0 mm and sintered at 1000°C-1400°C at 100°C interval for 10 hours. After final stage of sintering, samples estimated would shrink about 10% compared to its green body.

XRD were conducted to determine the crystalline structure of all samples by using Philips Panalytical X-Ray Diffractometer with Cu K$_\alpha$ radiation. The microstructure of the samples were analyzed by SEM (FEI Nov Nano SEM). The dielectric properties of the samples were determined using Agilent 4291B Impedance/material Analyzer in the microwave region in the frequency range 1 MHz to 1.5 GHz at room temperature.
RESULTS AND DISCUSSION

Figure 1 shows the XRD pattern of SrTiO$_3$ sintered at different temperatures of 1000°C, 1100°C, 1200°C, 1300°C and 1400°C, respectively. It showed ST samples were in crystalline structure. The small peaks fit the raw material SrCO$_3$. At 1200°C and above, no small peaks were found. The peak intensities of ST become more intense with increasing sintering temperature.

Figure 2 shows the SEM images of SrTiO$_3$ sintered at different temperatures: (a) 1000°C; (b) 1100°C; (c) 1200°C; (d) 1300°C; (e) 1400°C. It showed that there were agglomerations of powder bonded with grains. The powder is believed to be the raw materials that were not fully reacted until 1000°C. This may fit to the weak peaks in XRD patterns as discussed before. Necking of the grain can be obviously seen in Figures 2(c) - 2(d) at 1300°C and 1400°C respectively.
ST shrinkage percentage with respect to the sintering temperature is tabulated in Table 1. At 1000°C, there is no shrinkage of sample. There is a shrinkage of 0.25% at 1100°C. The shrinkage of the sample sintered at 1200°C increases to 5.25% from its original. At 1300°C, the shrinkage slightly decreases to 4.25%. At 1400°C, ST has the highest shrinkage percentage of 10.35% among other samples. From Figure 2(a) and Table 1, ST sintered at 1000°C, is said to have become coarsened. When coarsening process occurred, there is no net dimensional change (no shrinkage). The particles would only diffuse from surface to surface [9, 10].

Table 2 shows the average grain size of ST samples sintered at various temperatures. The average grain size increases with increasing sintering temperature from 0.1887 µm to 0.6647 µm. The average grain size of samples sintered at 1100°C and 1200°C is maintained at 0.25 µm. The increasing average grain size may be due to mass transport mechanism in the samples during the sintering process. As the sintering temperature increases, the driving force promoting neck growth increases [9]. Neck growth among the grains contributes to the increase in the average grain size.

Figure 3 shows the dielectric constant of ST samples sintered at 1000°C - 1400°C. The graph shows the same trend for all samples which is constant with increasing frequency. All samples except sample sintered at 1000°C show an increase in dielectric constant for frequency larger than 1 GHz. ST sintered at 1000°C has the lowest dielectric constant (~20). This may be due to incomplete perovskite phase formation and low crystallization of SrTiO$_3$ [11]. Another reason is that, it may also due to agglomerations of powder bonded with grains as mentioned above. Agglomerations of powder become a resistant for ions to polarize between grains and grain boundaries. ST sintered at 1200°C shows the highest dielectric constant of 50 among the samples. According to Teranishi et al [8], the dielectric constant of ST is only due to ionic polarization. The resonance effect is one of the reasons that caused dispersion in the dielectric constant at frequency less than 10$^7$ Hz [11]. From SEM images, the neck growth among the grains increases when sintering temperatures increases. On other hands, samples dielectric constant also possesses these phenomena. It is believed that, polarization of ions in grains may increased when grain necking growth. Polarization of ions in grains is a main factor to influences samples dielectric constant.

Figure 4 shows the dielectric loss of ST samples at room temperature with respect to frequency.
Samples sintered at 1200°C, 1300°C and 1400°C show the same trend with increasing frequency. All the samples showed relatively low loss, of less than 1 with frequency. The paraelectric perovskites show low losses in the GHz region due to ionic losses [12].

**CONCLUSIONS**

SrTiO₃ samples sintered at different temperatures were fabricated. The appearance of SrCO₃ in the XRD pattern shows incomplete formation of SrTiO₃ in samples sintered lower than 1200°C. Coarsening process occurred for sample sintered at 1000°C and it has no volume (%) shrinkage. Samples sintered at 1400°C have the highest shrinkage percentage of 10.35% among the samples. The average grain size increased with sintering temperature and it is due to neck growth among the grains. Neck growth among the grains contributes to increase the polarization of ions in grains, thus dielectric constant of samples is increasing when sintering temperature increases. ST samples sintered at 1200 °C exhibit the highest dielectric constant among the samples with a value of around 50. All the samples showed low loss which of less than 1 with frequency.

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