



UNIVERSITI PUTRA MALAYSIA

***THE INFLUENCE OF MICROSTRUCTURE EVOLUTION ON THE
DIELECTRIC PROPERTIES OF
STRONTIUM TITANATE AND ALUMINA SILICATE***

LEOW CHUN YAN

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By

LEOW CHUN YAN

**Thesis Submitted to the School of Graduate Studies, Universiti Putra
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Master of Science**

May 2015

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

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May 2015

Chair: Jumiah Hassan, PhD

Faculty: Science

Strontium Titanate (ST) and Alumina Silicate (AS) have wide range of applications as an advance ceramics. They are well known for their reliable performance especially at high temperature applications. Due to their amazing properties, most of the studies often focus on the after formation of its crystal structure phase and microstructure changes towards the dielectric properties. In fact, these studies are lacks of information link with the early stage of synthesis process. Wide range of sintering temperature and different particle size of green body must be set, in order to have a better understanding on the properties of ST and AS.

In this study, a series of these samples were prepared by conventional solid-state reaction method with two different particle sizes (sieved with laboratory test sieve of size 45 μm and 20 μm). These samples undergo a sintering process at temperatures from 500°C to 1400°C at 100°C intervals. Across this sintering temperature range. Average grain size of the samples were measured to investigate the microstructure evolution. Field Emission Scanning Electron Microscopy (FESEM) was carried out to capture samples microscopic images while X-ray diffraction (XRD) analysis was to study and confirmed the formation of crystal structure phase of ST and AS. Average grain size changes and crystal structure formation were studied in parallel with the dielectric properties at various measuring temperatures. Dielectric properties of ST and AS were analyzed at two different frequency range. For low frequency dielectric measurement, the frequency range is at 40 Hz – 1 MHz at measuring temperature from 28°C to 300°C with 50°C interval. For microwave dielectric measurements, the frequency range is at 1 MHz – 1.8 GHz at room temperature.

Based on XRD analysis, ST crystal structure started to form at 700°C sintering temperature. Synthesis was complete at 900°C. For AS, mullite compound was found and confirmed by XRD analysis at 1300°C and 1400°C sintering temperature. By observation, microstructure of ST started from agglomerated powder to form grains. As sintering temperature increases, grains continue to grow through the necking process. The evolution process is almost completed when grain boundaries diffuse with each other to form larger grains. By comparing both series of ST samples sintered at 1400°C, average grain size of the sample with greater starting particles size is 0.640 μm while another is 0.496 μm . ST45 (sieved with test sieve of 45 μm) was experiencing rapid grain growth at the final sintering stage, while ST20 (sieved with test sieve of 20 μm) grain growth was slower. For AS, thin and long rectangular shape grain structure were observed in samples sintered at 1400°C. ST samples with greater starting particles size form larger grain size.

At low frequency measurement, dielectric constant is dependent on frequency. Grain size will alter the dielectric constant of ST depending on the measuring temperatures. Loss tangent, $\tan \delta$ is also dependent on measuring temperature. Loss peak are strongly connected with frequency and measuring temperature. Agglomerations of powders will encourage alternating current conduction at high measuring temperature and causing the dielectric constant to drop. Dielectric constant of ST20 is greater than ST45 due to greater interfacial polarization effects occurred in ST20. ST20 has smaller grain size that would increase its surface effect. Rapid grain growth in ST45 had reduces its dielectric properties performances. Slower grain growth is important to promote homogeneous grain size and grains distribution during ST microstructure evolution. Homogeneous and smaller particle size are important factors to form high quality ST ceramic with good dielectric properties. For microwave frequency measurement, when frequency is beyond 10^7 Hz, dielectric constant value will remains constant across increasing frequency. Dielectric properties of ST is dependent of the grain size. ST with complete crystal structure will show greater dielectric constant value.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

PENGARUHAN EVOLUSI MIKROSTRUKTUR PADA SIFAT-SIFAT DIELEKTRIK BAGI STRONTIUM TITANAT DAN ALUMINA SILIKAL

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Strontium Titanat (ST) dan Alumina Silikal (AS) mempunyai pelbagai aplikasi sebagai seramik mara. Ia terkenal dengan prestasi mantap terutamanya dalam aplikasi suhu tinggi. Berdasarkan sifat-sifat mereka yang menakjubkan, kebanyakan penyelidikan selalu menumpu pada selepas pembentukan fasa struktur kristal dan perubahan mikrostruktur terhadap sifat dielektrik. Sebenarnya, penyelidikan-penyelidikan ini kekurangan maklumat berhubung dengan proses sintesis pada tahap awal. Suhu dengan jarak yang luas dan pelbagai saiz zarah badan hijau mesti ditetapkan untuk pemahaman yang lebih baik terhadap sifat-sifat ST dan AS.

Dalam kajian ini, sesiri sampel telah disediakan dengan kaedah reaksi konvensional keadaan pepejal dengan saiz zarah yang berbeza (disaring dengan ayak makmal ujian saiz 45 μm and 20 μm). Sample ini akan melalui proses penyepuhlingapan dari suhu 500°C ke 1400°C selang 100°C. Melalui jarak suhu penyepuhlingapan ini, purata saiz bijiran sampel telah diukur demi menyelidik evolusi mikrostruktur. Perlepasan medan mikroskop elektron imbasan (FESEM) telah dijalankan untuk mengambil imej mikroskopik sampel manakala pembelauan sinar-X (XRD) analisis adalah untuk mengkaji dan mengesahkan pembentukan fasa struktur kristal pada ST dan AS. Perubahan purata saiz bijiran dan pembentukan struktur kristal telah dikaji selari dengan sifat dielektrik pada suhu pengukuran yang berbeza. Sifat dielektrik bagi ST dan AS telah dianalisis pada jarak frekuensi yang berlainan. Untuk ukuran dielektrik frekuensi rendah, jarak frekuensi terletak pada 40 Hz – 1 MHz dengan suhu pengukuran dari 28°C to 300°C selang 50°C. Manakala bagi ukuran dielektrik gelombangmikro, jarak frekuensi terletak pada 1 MHz – 1.8 GHz dalam suhu bilik.

Berdasarkan XRD analisis, struktur kristal ST mula membentuk pada 700°C suhu penyepuhlingapan. Sintesis telah dilengkapkan pada 900°C suhu penyepuhlingapan. Untuk AS, kompaun mullite telah dijumpa dan disahkan dengan analisis XRD pada 1300°C dan 1400°C suhu penyepuhlingapan. Dari pemerhatian, evolusi pada ST

bermula membentuk bijiran dari gumpalan serbuk. Dengan peningkatan suhu penyepuhlindapan, bijiran terus mengembang melalui proses membentuk leher. Proses evolusi hampir menamat apabila sempadan bijiran meresap antara satu sama lain bagi membentuk bijiran yang lebih besar. Dengan perbandingan dua siri sampel ST yang disinter pada 1400°C, purata saiz bijiran oleh sampel dengan permulaan saiz zarah yang lebih besar adalah 0.640 μm manakala yang satu lagi adalah 0.496 μm . ST45 (disaring dengan ayak ujian 45 μm) mengalami pembentukan bijiran yang pesat pada peringkat akhir penyepuhlindapan manakala pembentukan bijiran ST20 (disaring dengan ayak ujian 45 μm) lebih perlahan. Bagi AS, bijiran bentuk jalur telah diperhatikan dalam sampel yang disinter pada 1400°C. Sample ST dengan permulaan saiz zarah yang lebih besar akan membentuk bijiran yang lagi besar.

Pada ukuran frekuensi rendah, pemalar dielektrik bergantung kepada frekuensi. Saiz bijiran akan menukarkan pemalar dielektrik ST yang bergantung pada suhu pengukuran. Tangen kehilangan, $\tan \delta$ bergantung pada suhu pengukuran. Puncak kehilangan adalah amat berkaitan dengan frekuensi dan suhu pengukuran. Gumpalan serbuk akan menggalakan pengaliran arus ulang alik pada suhu pengukuran yang tinggi dan menurunkan nilai pemalar dielektrik. Nilai pemalar dielektrik ST20 adalah lebih besar dari ST45 kerana kesan pengutupan antara muka yang lebih kuat belaku dalam ST20. ST20 mengandungi saiz bijiran yang kecil yang dapat meningkatkan kesan permukaan. Akibat pembentukan bijiran yang pesat dalam ST45 telah mengurangkan prestasi sifat dielektriknya. Sifat dielektrik ST bergantung kepada saiz bijiran. Kadar pembentukan bijiran yang perlahan adalah amat penting untuk menggalakan saiz bijiran yang homogen dan taburan bijiran semasa evolusi mikrostruktur ST. Saiz zarah yang lebih kecil dan homogen merupakan faktor-faktor amat penting bagi membentuk seramik ST yang bermutu tinggi dengan sifat dielektrik yang bagus. Bagi pengukuran pada frekuensi gelombangmikro, apabila frekuensi telah melebihi 10^7 Hz, nilai pemalar dielektrik akan tetap sepanjang peningkatan frekuensi. Sifat dielektrik ST bergantung kepada saiz bijiran. ST dengan struktur kristal yang lengkap akan menunjuk nilai pemalar dielektrik yang lebih besar.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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LIST OF ABBREVIATIONS

C	Capacitance
G	Conductance
f	Frequency
E	Electric Field
σ	Magnitude of Surface Charge Density
Q	Magnitude of Total Charge
V	Potential Difference
A	Area of Each Plate / Area of sample
d	Separation between the plates
C_{vac}	Capacitance of Parallel Plates in Vacuum
ϵ_0	Permittivity of Free Space
ϵ^*	Complex Permittivity
ϵ_r^*	Relative Complex Permittivity
j	Imaginary Operator with $(-1)^{1/2}$
ϵ_r'	Relative Dielectric Constant
ϵ_r''	Relative Dielectric Loss Factor
δ	Loss angle
$\tan \delta$	Dielectric Loss Tangent
n	Order of the Diffracted Beam
λ	Wavelength of the Incident X-Ray Beam
hkl	Lattice Plane
d_{hkl}	Spacing Between atomic planes
θ_{hkl}	Angle of the Incident X-Ray
$^\circ$	Degree
$^\circ\text{C}$	Degree Celsius
Hz	Hertz

Short Form	Description
Al	Aluminum
Ba	Barium
Ca	Calcium
O	Oxygen
Sr	Strontium
Ta	Tantalum
Ti	Titanium
Zr	Zirconium
Al ₂ SiO ₅	Alumina Silicate
Al ₂ O ₃	Aluminum Oxide
SiO ₂	Silicon Oxide
SrTiO ₃	Strontium Titanate
SrCO ₃	Strontium Carbonate
SrO ₂	Strontium Peroxide
Sr(NO ₃) ₂	Strontium Nitrate
BaTiO ₃	Barium Titanate
N ₂	Nitrogen Gas
Fe ₂ O ₃	Iron Oxide
ac	Alternating Current
AS	Alumina Silicate
DRAM	Dynamic Random Access Memories
FESEM	Field Emission Scanning Electron Microscope
GBLC	Grain Boundary Layer Capacitor
ICDD	International Centre for Diffraction Data
SEM	Scanning Electron Microscope
T _c	Curie temperature
XRD	X-ray Diffraction
YBCO	Y-Ba-Cu-O Superconductor

CHAPTER 1

INTRODUCTION

1.1 Ceramics

Typical ceramics may be defined as inorganic non-metallic material, which occur in crystalline structure, partial crystalline structure or amorphous form. Ceramics offer many advantages compared to other materials. Comparing with other materials, ceramics are far more chemically inert and durable. Besides, they also demonstrate excellent strength and hardness properties. They are harder and stiffer than steel. Ceramics do not deform easily at ordinary temperatures (Tilley, 2004). Due to its outstanding chemical inert properties, ceramics are more corrosion resistant than metals or polymers. Although with all kinds of pros as mentioned, ceramics does have some cons in nature. Generally, ceramics are brittle and easy to fracture under impact. However, even ceramics are readily fractured when stretched; they are much stronger when compressed.

Ceramic is an important material throughout the history of humankind. Even until today and in the future, ceramic is still has it crucial roles to play in modern technology. People may underestimate the potential applications of ceramics because they believe ceramics are all about pottery and tiles. In fact, besides this kind of traditional use, ceramic also possess outstanding properties that are suitable for modern technology especially in electronic industries. The evolution from pottery to electronic components has broadened the term 'ceramic' (Moulson et.al., 1990). Ceramic materials display a wide range of properties that facilitate their use in many different potential areas. Advanced ceramic has different functions in different fields. Among the fields are electrical, magnetic optical, biomedical and many others. In electrical field, applications of ceramic are broad. Ceramic BaTiO_3 and SrTiO_3 are representative in capacitors and microwave dielectrics. For superconductors is $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO). Ceramics are also used as piezoelectric material, electrical insulator and solid-oxide fuel cell that are under electrical field (Askeland et. al., 2003).

Ferrite-type and garnet-type ceramics are representative in magnetic field. Magnets, inductors (choke), circulators and isolators mostly are made from ferrite and garnet type ceramics. They are the core in transformers and microwave technology (Askeland et. al., 2003; Moulson et.al., 1990). SiO_2 and Al_2O_3 based ceramics mostly function in optical field. Normally they are made into glasses and as based material in lasers. Under biomedical field, ceramics serve as implants such as dental materials. Finally, yet importantly is the contribution of ceramic in construction and domestic application, for examples: concretes, tiles, sanitary wares and kitchenwares. With all the functions and application of ceramic, it is like a great gift from the creator of the world. We should not take it for granted and must use it wisely.

1.2 Strontium Titanate, SrTiO₃

Strontium Titanate (ST) or Tausonite named to honour Lev Vladimirovich Tauson (1917–1989), a Russian geochemist petrologist. ST with chemical formula SrTiO₃ is an oxide of Strontium and Titanium. In nature, ST mineral is extremely rare and usually it occurred in a form of very tiny crystal. Almost all ST use in application is synthetically prepared by raw materials rather than from ore extractions.

ST may occur in brown, red-brown, grey and dark grey colour. Its Mohs Hardness value is around 6.0 – 6.5 while for diamond is 10. As a typical ceramic material, ST is brittle and may show conchoidal fracture when it breaks. Conchoidal fracture is the way it breaks that do not follow any natural planes of separation. Theoretical density of ST is 4.81 g cm⁻³. Its melting point is 2080 °C. At room temperature, ST has dielectric constant around 330 and dielectric loss of the order of 10⁻³ (Jacob et al., 2011).

Application of ST is mostly found in electronic industries. ST has no phase transformation above the ferroelectric - paraelectric transition temperature, T_c. It possessed excellent stable dielectric properties and stable temperature characteristics (Zhao et al., 2004). With these excellent properties, ST is suitable for use in grain boundary layer capacitor (GBLC) and as thin film in dynamic random access memories (DRAMs) (Roy et al., 2005). ST may also become a good microwave tunable device in situations of dependence in high dc electric field (Amaral et al., 2009). Besides, ST exhibits nonlinear current–voltage (I–V) characteristic that is suitable to be used as varistor (Li et al., 2006). Furthermore, ST is also important in other applications such as oxygen gas sensor, diamond stimulant and as substrate for the hetero-epitaxial growth of high T_c-superconductors (Wang et al., 2008; Roy et al., 2005). Since ST has the outstanding characteristic in its applications, until today it is still a worthy material to research and study.

Seeing that ST has such excellent characteristic in a wide range of applications, it aroused many researchers interest and attention, to study what is the reason that made ST such an outstanding material. From material scientist point of view, macroscopic properties of a material may always be affected by its structure at atomic or molecular scales. ST appeared in perovskite structure form. Perovskite is a family name of ABO₃ compound group where A and B differ in size. Normally, A is a large radius cation of coordinate 12 (example: Ba, Sr, Ca, Pb) which occupies the empty sites between oxygen (O) octahedral. B will be a smaller radius cation of coordinate 6 (example: Ti, Zr, Nb, Ta) (Bunget et. al., 1984).

At room temperature, ST is in cubic perovskite structure with space group *Pm3m* (Shkabko et al., 2009). In the structure, Oxygen (O) ions formed octahedron structure lies in the body- and face-centred cubic structure formed by A-Strontium (Sr) and B-Titanium (Ti) ions (Figure 1.1). SrTiO₃ compound with this structure possessed paraelectric characteristic. Either the distortions of the oxygen octahedra gathering or decentration distortion of ion B in oxygen octahedra may dramatically change the characteristic of the materials. For example, Barium Titanate (BaTiO₃) has distorted B

ions (Ti) at room temperature that makes it become tetragonal structure. Displacement of B ions (Ti) with O ions changed the interatomic bonding forces that increase the covalency of B-O bond. This interesting phenomenon has made Barium Titanate a famous ferroelectric material (Bunget et., al. 1984).

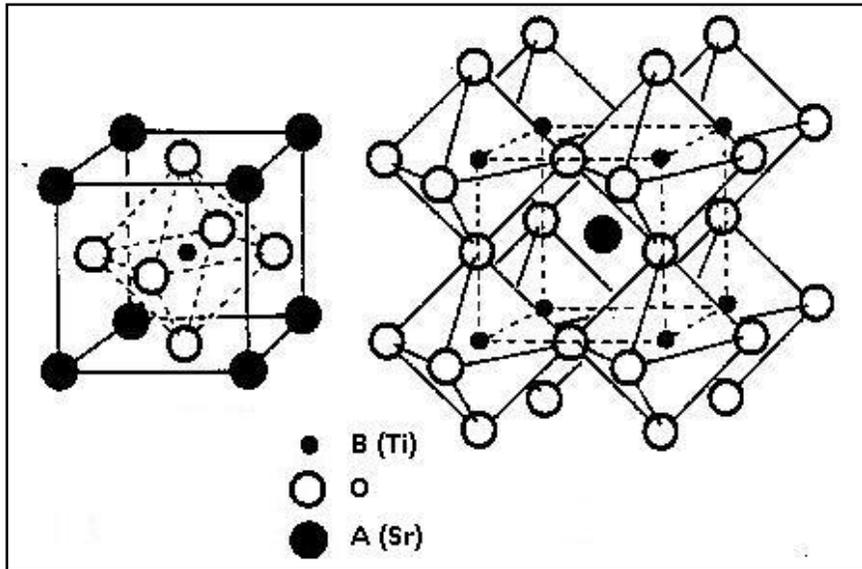


Figure 1.1 Cubic Perovskite Structure of ST (Xu,1991)

1.3 Alumina Silicate, Al_2SiO_5

Alumina Silicate or Aluminum Silicate (AS) mainly is formed by mixture of Aluminium (Al), Silica (Si) and Oxygen (O). AS exist in several crystal structure forms by nature or depending on the synthesis condition. During synthesis, under different conditions of pressure and temperature, different crystalline forms and phase may occur. They could be kyanite, andalusite, sillimanite, mullite, kaolinite (combination of AS with water) and others. Kyanite, sillimanite and andalusite are the polymorphs occurring in AS minerals (Whitney, 2002). Kyanite forms in a lower temperature/higher pressure environment, andalusite forms in a lower temperature/lower pressure environment, while sillimanite forms in a higher temperature/higher pressure environment. For mullite, the molecular ratio is $3Al_2O_3 \cdot 2SiO_2$. Mullite rarely occur as a mineral in nature. It is commonly formed by synthesis. Chemical formula for all of these crystalline forms and phase is Al_2SiO_5 . Although they have the same chemical formula, they do exhibit some common and unique physical and chemical properties.

Most of AS ceramics have common properties of high melting point (Baker et. al., 2006), good thermal and chemical stability (Naga et. al., 1992). With excellent high temperature properties, AS products normally would serve as refractory material, fire

protection, metal melting and engineering applications. (Laskowski et. al., 1994; Baker et. al., 2006)

In this work, mullite is the material to be studied. Mullite a promising material that widely being use in traditional and advanced ceramics. It is outstanding with electrical, thermal and mechanical properties. The advantageous properties of mullite include high melting point, high temperature strength, excellent creep and chemical resistance, strong heat and electrical insulation and finally yet importantly are its low thermal expansion coefficient (Ebadzadeh et. al., 2009; Sainz et. al., 2000). Low loss in dielectric, good thermal and chemical stability allow mullite to perform as advanced ceramic for use in electronic packaging materials (thermal and electronic insulation purpose). Besides, mullite fibers are used for coating and even as a matrix to enhance composite materials. As a traditional ceramics, mullite is use as white wares, structural materials and refractory materials (Chen and Lan, 2000; Schneider et. al., 2005).

Mullite is orthorhombic - dipyramidal class with space group $Cmmm$. The average structure of mullite can be derived from the closely related but structurally simpler sillimanite. In sillimanite, like in other mullite type structures, edge-connected AlO_6 octahedral chains run parallel to the crystallographic c-axis in Figure 1.2. In sillimanite these octahedral chains are cross-linked by double chains with alternating AlO_4 and SiO_4 tetrahedra (Figure 1.2). Mullite can be derived from sillimanite by a coupled substitution, according to $2Si^{4+} + O^{2-} \rightarrow 2Al^{3+} + X$, $X =$ oxygen vacancy. This reaction involves removal of oxygen atoms from the structure leading to oxygen vacancies and to a rearrangement and disordering of tetrahedral cations (Schneider et. al., 2008; Schneider et. al., 2005). Understanding the structure of mullite is an important key to study its excellent properties.

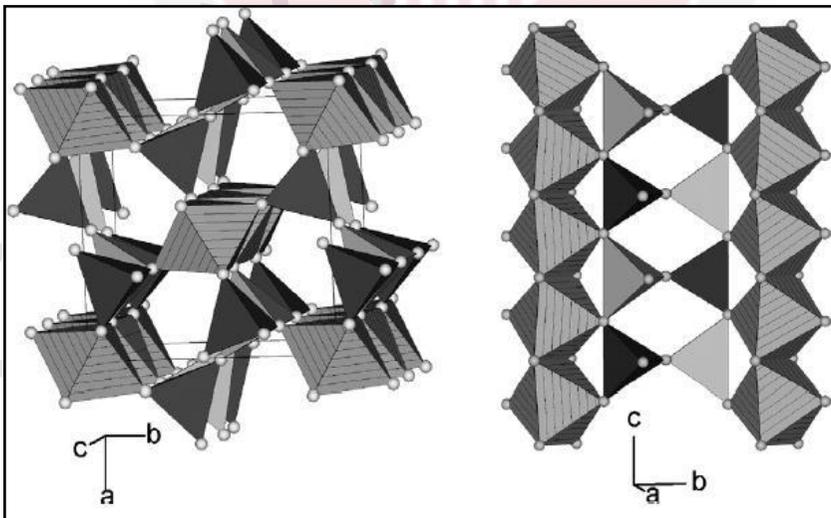


Figure 1.2: Crystal Structure of Sillimanite in projection down c axis and a axis (Simple model of Sillimanite and Mullite are alike) (Schneider et. al., 2008)

1.4 Background and importance of studies

Recent reports about Strontium Titanate (ST) and Alumina Silicate (AS) rarely studied on their dielectric properties across wide range of sintering temperatures. There are very few reviews made on a detailed study relating them from initial stage to final stage of sintering along with the microstructure and the dielectric properties are evolving towards their final form and values. Most of the studies often focus on the after formation of its crystal structure phase and microstructure changes towards the dielectric properties. In fact, these studies are lacks of information link with the early synthesis process.

The present research proposal intends to deal with these little understood aspects. In this study, ST and AS microstructure evolution were analyzed in parallel with their dielectric properties along with wide range sintering temperatures from 500°C to 1400°C. Microstructure changes in a ceramic material will always influence its application properties in parallel. As for sintering process, often, average grain size of a sample is one of the important key to unlock the information about microstructure evolution. In order to have a glance on microstructure evolution, grain size analysis is necessary to be studied.

Besides grain size analysis, X-Ray Diffraction (XRD) analysis needed to be carried out in order to study and confirmed the formation of crystal structure phase of ST and AS. Across wide range of sintering process, crystal structure formation needed to be studied along with the microstructure evolution to get a clearer picture on how the sintering temperature might affects the process. XRD analysis might provide more information to support the microstructure analysis.

Materials microstructure formation also is dependent on the parameters of its starting materials. Particle size of a starting material is one of the important parameter to influence the microstructure formation in most of the ceramic materials. Conventional solid-state reaction methods are the simplest and most widely used method to synthesize ceramics. In order to study the particular effects of starting materials particle size for ST, two different sieving size were set during solid-state reaction method was carried out. Different particle sizes of starting material will have yield different outcome for the microstructure morphology and they might influences the dielectric properties as well.

Dielectric measurements are performed at frequencies from 40 Hz to 1.8 GHz. Within this frequency range, it is the optimum condition to analyse the dielectric properties of the samples for application. Strontium Titanate and Alumina Silicate are well known for their stable dielectric properties performance at temperature above 100°C. Various measuring temperature were set parallel during the dielectric measurement is being carried out. From this study, the optimization process for microstructure phase formation may lead to optimize its dielectric properties. Analysis of the data obtained can be extended to other alumina and titanate based materials for the development of multilayer capacitors.

1.5 Objectives

Strontium Titanate and Alumina Silicate samples are synthesized at sintering temperature range from 500°C - 1400°C. Across this sintering temperature:

1. Samples preparation were set with two different particle sizes using conventional solid-state reaction method. This setting is made in order to investigate the fundamental core that influence the microstructure formation.
2. Crystal structure formation of samples are analyzed by XRD analysis and average grain size of samples are determined from the Scanning Electron Microscopy (SEM) micrograph. Both these methods will monitored the crystal structure formation and microstructure evolution along with increasing sintering temperature.
3. Dielectric properties of samples are studied from 40 Hz to 1.8 GHz frequency range with different measuring temperature. Relationship between the dielectric properties and microstructure of the samples were studied in parallel in order to have a better and deeper understanding on this mutual dependent parameters.

1.6 Limitations

1. All samples are only prepared by conventional solid state reaction method and the particle sizes are limited at micron size range.
2. Sintering temperature of AS samples are limited only to 1400°C due to furnace limitation (no furnace higher than 1400°C was available).

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