



UNIVERSITI PUTRA MALAYSIA

**DIELECTRIC PROPERTIES OF STRONTIUM TITANATE AND
CALCIUM TITANATE PREPARED VIA SOLID STATE
AND MECHANICAL ALLOYING METHODS**

WONG YICK JENG

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**DIELECTRIC PROPERTIES OF STRONTIUM TITANATE AND CALCIUM
TITANATE PREPARED VIA SOLID STATE AND MECHANICAL
ALLOYING METHODS**

By

WONG YICK JENG

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,
in Fulfilment of the Requirements for the Degree of Master of Science**

April 2013



SPECIALLY DEDICATED TO

- My kind and helpful supervisor and co-supervisor, Associate Professor Dr. Jumiah Hassan and Associate Professor Dr. Mansor Hashim
- My beloved family for their endless support and encouragement
- My seniors and all my friends for their advice and assistance

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

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Chairman: Jumiah Hassan, PhD

Faculty: Science

In this thesis, the phase formation, microstructure, and dielectric properties of strontium titanate (SrTiO_3) and calcium titanate (CaTiO_3) were investigated. Both samples were prepared using two different techniques, one by solid state reaction technique with the sintering temperature from 1200 to 1400°C at 40°C interval, and another by mechanical alloying method with the sintering temperature from 1000 to 1200°C at 40°C interval. From x-ray diffraction (XRD) analysis at room temperature, a single phase cubic SrTiO_3 structure with space group $Pm-3m$ and a single phase orthorhombic CaTiO_3 structure with space group $Pnma$ were obtained at 1200°C and above as both samples were prepared by solid state method. For the SrTiO_3 samples prepared by mechanical alloying method, the formation of a single phase cubic SrTiO_3 structure with space group $Pm-3m$ could be achieved at 1160°C and above; whilst, a single phase orthorhombic CaTiO_3 with space group $Pnma$ could be formed starting from 1120°C and above. In all the synthesized samples, the morphological

analysis revealed that the grains were in submicron range with the calculated average grain sizes increased with sintering temperature.

The real part of relative permittivity ϵ_r' , imaginary part of relative permittivity ϵ_r'' , and loss tangent $\tan \delta$ have been measured on the sintered samples with respect to frequency range 0.01 Hz-1 MHz and varying temperatures from 25 to 250°C at 25°C interval. Meanwhile, the microwave dielectric measurements in the frequency range 1 MHz-1.5 GHz were also performed at room temperature. In the dielectric analysis at low frequency, ϵ_r' decreased with increasing frequency for both samples prepared by solid state and mechanical alloying methods. The values of ϵ_r' and $\tan \delta$ showed an increase with the rise in temperature. The loss of all the samples under study at different measured temperatures were characterized by the loss peaks at particular frequencies. It indicated the presence of conductivity relaxation in the material. In the microwave frequency range, ϵ_r' remained constant for the SrTiO₃ and CaTiO₃ prepared by both methods, notably in the frequency range 10⁷-10⁸ Hz, whereas the values of ϵ_r'' were relatively small which lied in the order of 10⁻² in the same frequency range. At 1200°C, the room temperature values of ϵ_r' at 1 and 10 MHz of solid state prepared SrTiO₃ sample were about 27.7 and 43.6 respectively, while the room temperature values of ϵ_r' of mechanical alloyed SrTiO₃ samples were about 58.8 and 65.7 respectively. The room temperature values of ϵ_r' at 1 and 10 MHz for the solid state prepared CaTiO₃ samples sintered at 1200°C were about 19.4 and 42.4 respectively, while were about 132 and 205 respectively for the mechanical alloyed CaTiO₃ samples.

For all the investigated samples, ϵ_r' values increased with increasing sintering temperature in the low and microwave frequency range due to the grain growth with

respect to increasing sintering temperature. The grain growth rate of mechanical alloyed samples was larger than that of solid state prepared samples. Hence, the values of ϵ_r' were found to be enhanced in the samples prepared by mechanical alloying method. It was thus concluded that mechanical alloying processing route was thus a good alternative and yielded materials with improved dielectric properties if compared to that solid state method.



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

**SIFAT DIELEKTRIK BAGI STRONTIUM TITANAT DAN KALSIUM
TITANAT DISEDIAKAN MELALUI KAEDAH PEMROSESAN SERAMIK
DAN PENGALOIAN MEKANIKAL**

Oleh

WONG YICK JENG

April 2013

Pengerusi: Jumiah Hassan, PhD

Faculti: Sains

Dalam tesis ini, pembentukan fasa, mikrostruktur, dan sifat dielektrik bagi strontium titanat (SrTiO_3) dan kalsium titanat (CaTiO_3) telah dikaji. Kedua-dua sampel telah disediakan dengan menggunakan dua teknik yang berlainan, satu melalui teknik pemrosesan seramik disinter dari suhu penyepuhlindapan 1200 ke 1400°C dengan selang 40°C, dan satu lagi disediakan melalui cara pengalioian mekanikal disinter dari suhu penyepuhlindapan 1000 ke 1200°C dengan selang 40°C. Dari analisis XRD pada suhu bilik, fasa tunggal bagi struktur kubus SrTiO_3 dengan kumpulan ruang $Pm-3m$ dan fasa tunggal bagi struktur ortorombus CaTiO_3 dengan kumpulan ruang $Pnma$ telah didapati pada suhu 1200°C ke atas bagi kedua-dua sampel yang disediakan dengan menggunakan pemrosesan seramik. Bagi sampel SrTiO_3 yang disediakan dengan menggunakan cara pengalioian mekanikal, pembentukan fasa tunggal bagi struktur kubus SrTiO_3 dengan kumpulan ruang $Pm-3m$ boleh dicapai pada suhu 1160°C ke atas, manakala fasa tunggal bagi struktur ortorombus CaTiO_3 dengan kumpulan ruang $Pnma$ boleh dibentuk bermula dari suhu 1120°C ke atas.

Dalam semua sampel tersinter, analisis morfologi mendedahkan butiran adalah dalam rantau submikron dengan purata saiz bijian menunjukkan peningkatan terhadap suhu penyepuhlingapan.

Ketelusan nyata relatif ϵ_r' , ketelusan khayalan relatif ϵ_r'' , dan tangen kehilangan $\tan \delta$ telah diukur terhadap sampel yang telah disinter dari frekuensi 0.01 Hz ke 1 MHz dan juga berubah terhadap pemanasan suhu dari 25 ke 250°C dengan selang 25°C. Sementara itu, frekuensi mikrogelombang dari 1 MHz ke 1.5 GHz juga dijalankan pada suhu bilik. Dari analisis dielektrik pada frekuensi rendah, ϵ_r' menurun dengan peningkatan frekuensi bagi kedua-dua sampel yang disediakan menggunakan pemprosesan seramik dan cara pengaloiian mekanikal. Nilai untuk ϵ_r' dan $\tan \delta$ menunjukkan peningkatan dengan peningkatan dalam pemanasan suhu. Kehilangan dielektrik bagi semua sampel yang dikaji pada pelbagai pemanasan suhu dicirikan oleh puncak $\tan \delta$ pada frekuensi yang tertentu. Ini menandakan kewujudan santonian keberaliran dalam sampel. Dalam frekuensi mikrogelombang, ϵ_r' adalah malar bagi SrTiO₃ dan CaTiO₃ sampel yang disediakan menggunakan kedua-dua teknik, terutama dari frekuensi 10⁷ ke 10⁸ Hz, manakala nilai bagi ϵ_r'' adalah agak kecil di mana ϵ_r'' berkumpul di aras 10⁻² di antara frekuensi yang sama. Pada suhu 1200°C, nilai untuk ϵ_r' yang diukur pada 1 dan 10 MHz di suhu bilik bagi SrTiO₃ yang disediakan menggunakan pemprosesan seramik ialah 27.7 dan 43.6 masing-masing, manakala untuk ϵ_r' bagi SrTiO₃ yang disediakan menggunakan cara pengaloiian mekanikal pada suhu bilik ialah 58.8 dan 65.7 masing-masing. Nilai untuk ϵ_r' yang diukur pada 1 dan 10 MHz di suhu bilik bagi CaTiO₃ yang disediakan menggunakan pemprosesan seramik ialah 19.4 dan 42.4 masing-masing, manakala nilai-nilai untuk ϵ_r' bagi CaTiO₃ yang disediakan menggunakan cara pengaloiian mekanikal pada suhu bilik ialah 132 dan 205 masing-masing.

Bagi semua sampel yang dikaji, nilai bagi ϵ_r' meningkat dengan peningkatan suhu penyepuhlindapan pada frekuensi rendah dan frekuensi mikrogelombang disebabkan penumbuhan butiran terhadap peningkatan suhu penyepuhlindapan. Kadar penumbuhan butiran bagi sampel yang disediakan menggunakan cara pengaloiian mekanikal adalah besar berbanding dengan sampel yang disediakan melalui pemprosesan seramik. Oleh itu, nilai bagi ϵ_r' didapati meningkat bagi sampel yang disediakan melalui cara pengaloiian mekanikal. Maka ini menyimpulkan bahawa teknik pengaloiian mekanikal ialah alternatif yang bagus dan menghasilkan bahan-bahan dengan sifat dielektrik yang meningkat jika dibandingkan dengan pemprosesan seramik.

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I certify that a Thesis Examination Committee has met on 25 April 2013 to conduct the final examination of Wong Yick Jeng on his thesis entitled “Dielectric Properties of Strontium Titanate and Calcium Titanate Prepared via Solid State and Mechanical Alloying Methods” in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the degree of Master of Science.

Members of the Thesis Examination Committee were as follows:

Azmi Zakaria, PhD

Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Abdul Halim Bin Shaari, PhD

Professor
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Lim Kean Pah, PhD

Senior Lecturer
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Ibrahim Abu Talib, PhD

Professor
Faculty of Science and Technology
Universiti Kebangsaan Malaysia
Malaysia
(External Examiner)

NORITAH OMAR, PhD

Associate Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 26 June 2013

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

Jumiah Hassan, PhD

Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Mansor Hashim, PhD

Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

Wan Daud Wan Yusoff, PhD

Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

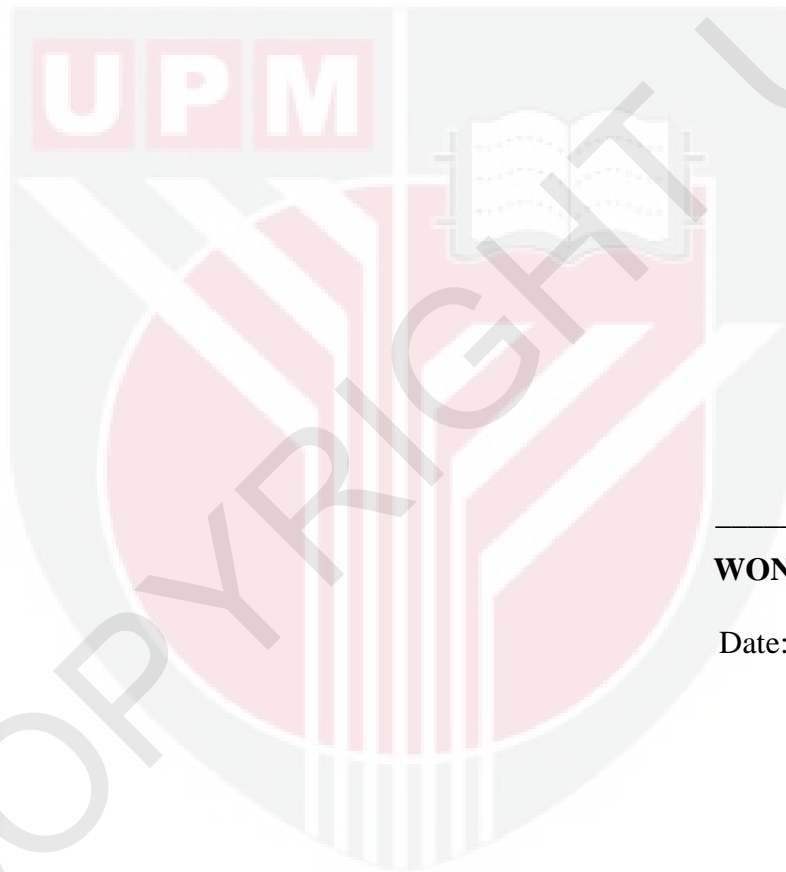
BUJANG BIN KIM HUAT, PhD

Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.



WONG YICK JENG

Date: 25 April 2013

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