



UNIVERSITI PUTRA MALAYSIA

**THERMAL DIFFUSIVITY AND DIELECTRIC PROPERTIES OF
STRONTIUM- DOPED BARIUM TITANATE AND CALSIUM TITANATE
SYSTEMS.**

NOOR JAWAD RIDHA

FS 2009 12



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**MASTER OF SCIENCE
UNIVERSITI PUTRA MALAYSIA**

2009



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By

NOOR JAWAD RIDHA

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

April 2009



DEDICATION

To my father soul

To my dearest mother
for her supports and prayer.....

To my husband Firas Kamel Mohamad
who loves and support me all the times...

To my supervisors
Prof. Dr. W. Mahmood Mat Yunus,
Prof. Dr. Abdul Halim Shaari and Assoc. Prof. Dr. Zainal Abidin Talib
for their time and advice

To all my friends
for their assistance and supports...

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

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April 2009

Chairman: W. Mahmood Mat Yunus, PhD

Faculty: Science

In this thesis, the structure, thermal diffusivity and dielectric constant of $Ba_{1-x}Sr_xTiO_3$ and $Ca_{1-x}Sr_xTiO_3$ ($0 \leq x \leq 1$) ceramics were investigated. The samples were prepared using solid-state reaction technique with a sintering temperature at 1200 °C. From XRD analysis, the $BaTiO_3$ structure obtained was tetragonal and then transformed to cubic; whilst, $CaTiO_3$ structure changed from orthorhombic to cubic with an intermediate tetragonal phase as the amount of Sr ions increased. Surface morphology studies showed that the grain size decreased with increasing Sr ions in both $Ba_{1-x}Sr_xTiO_3$ and $Ca_{1-x}Sr_xTiO_3$ systems.

Photoflash technique was used to determine the thermal diffusivity of $BaTiO_3$, $CaTiO_3$ and $SrTiO_3$ at room temperature. The effect of substitution Sr ions on the thermal diffusivity of $Ba_{1-x}Sr_xTiO_3$ and $Ca_{1-x}Sr_xTiO_3$ was also investigated. It was found that increasing Sr ions in $Ba_{1-x}Sr_xTiO_3$ samples reduced the thermal diffusivity value from $11.302 \times 10^{-3} \text{ cm}^2/\text{s}$ to $6.467 \times 10^{-3} \text{ cm}^2/\text{s}$ and accompanying by a decrease in density. Similarly, the thermal diffusivity values of $Ca_{1-x}Sr_xTiO_3$ system decreased from $13.11 \times 10^{-3} \text{ cm}^2/\text{s}$ to $6.467 \times 10^{-3} \text{ cm}^2/\text{s}$ as its density increased.



For thermal diffusivity measurement at higher temperature, laser flash technique was used. It was noticed that the thermal diffusivity of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ and $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$ decreased with increasing temperature from room temperature to 150 °C.

The dielectric properties of BaTiO_3 , CaTiO_3 and SrTiO_3 were investigated at various temperatures from 25 to 150 °C using AC impedance analyzer. Increasing Sr ions in BaTiO_3 reduced the dielectric constant from 709 to 246 at frequency 10^6 Hz at room temperature. On the other hand, increasing Sr ions in CaTiO_3 raised the dielectric constant from 106 to 246, and the highest value was found at $x = 0.2$. The dielectric constant of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ decreased with increasing temperature. The highest dielectric constant value was recorded for $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ system where $x = 0$ and 0.1 at phase change temperatures, 125 °C and 100 °C respectively.

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

**RESAPAN TERMA DAN SIFAT DIELEKTRIK
BARIUM TITANATE DAN KALASIAM TITANATE
DIDOP DENGAN STRONTIUM**

Oleh

NOOR JAWAD RIDHA

April 2009

Pengerusi: W. Mahmood Mat Yunus, PhD

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Dalam tesis ini, struktur, resapan terma dan pemalar dielektrik bagi seramik $Ba_{1-x}Sr_xTiO_3$ dan $Ca_{1-x}Sr_xTiO_3$ ($0 \leq x \leq 1$) telah dikaji. Sampel telah disediakan dengan menggunakan teknik tindak balas pepejal dan disinter pada suhu $1200^\circ C$. Daripada analisis XRD, struktur $BaTiO_3$ didapati berubah dari tetragonal kepada struktur kubus, manakala bagi struktur $CaTiO_3$, didapati berubah dari ortorombik kepada struktur kubus dengan pertengahan fasa tetragonal apabila ion Sr bertambah. Kajian morfologi permukaan menunjukkan bahwa saiz butiran mengecil dengan meningkatnya Sr ions di dalam kedua- dua sistem $Ba_{1-x}Sr_xTiO_3$ dan $Ca_{1-x}Sr_xTiO_3$.

Teknik fotokilat telah digunakan untuk menentukan resapan terma $BaTiO_3$, $CaTiO_3$ dan $SrTiO_3$ pada suhu bilik. Kesan penggantian ion Sr terhadap respon terma dalam $BaTiO_3$ dan $CaTiO_3$ juga telah dikaji. Didepeti bahawa pertambahan ion Sr di dalam sampel $BaTiO_3$ mengurangkan nilai kadar resapan terma dari $11.302 \times 10^{-3} \text{ cm}^2/\text{s}$ kepada $9.6467 \times 10^{-3} \text{ cm}^2/\text{s}$, dan diikuti dengan penurunan dalam ketumpatan.

Serupa juga, nilai resapan terma sistem $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$ berkurangan dari $13.11 \times 10^{-3} \text{ cm}^2/\text{s}$ kepada $6.467 \times 10^{-3} \text{ cm}^2/\text{s}$, sebaliknya nilai ketumpatan bertambah.

Untuk pengukuran resapan terma pada suhu yang lebih tinggi, teknik kilatan laser telah digunakan. Didapati bahawa resapan terma bagi $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ dan $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$ menurun dengan kenaikan suhu dari 25°C hingga 150°C .

Sifat dielektrik BaTiO_3 , CaTiO_3 dan SrTiO_3 diukur pada belbagai suhu dalam julat 25°C hingga 150°C dengan menggunakan penganalisis impedans. Pertambahan ion Sr dalam BaTiO_3 telah mengurangkan pemalar dielektrik (pada 10^6 Hz) dari 709 kepada 246. Sementara itu, pertambahan ion Sr dalam CaTiO_3 meningkatkan pemalar dielektrik dari 106 kepada 246, dan nilai tertinggi dicapai pada $x = 0.2$. disamping itu, pemalar dielektrik bagi sistem $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ dengan $x = 0$ dan 0.1 didapati pada suhu perubahan fasa 100°C dan 125°C .

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I certify that an Examination Committee has met on 28 April 2009 to conduct the final examination of Noor Jawad Ridha on her Master of Science thesis entitled “Thermal Diffusivity and Dielectric Constant of Strontium- Doped Barium Titanate and Calcium Titanate Systems” in accordance with Universiti Putra Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the student be awarded the degree of Master of Science.

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DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

**NOOR JAWAD
RIDHA**

Date: 20 May 2009

TABLE OF CONTENTS

	Page
DEDICATION	ii
ABSTRACT	iii
ABSTRAK	v
ACKNOWLEDGEMENTS	vii
APPROVAL	viii
DECLARATION	x
LIST OF TABLES	xiv
LIST OF FIGURES	xv
LIST OF SYMBOLS	xviii
LIST OF ABBREVIATIONS	xxi
 CHAPTER	
1	INTRODUCTION 1
	1.1 Introduction to the Ceramics and its Applications 1
	1.2 Ferroelectric Materials 3
	1.3 Phase Transitions 5
	1.4 Thermal Diffusivity 5
	1.5 Research Problems 7
	1.6 Importance of the Study 8
	1.7 Objectives of the Study 8
	1.8 Outline of the Present Work 9
2	LITERATURE REVIEW 10
	2.1 Introduction 10
	2.2 Various Techniques to Measure Thermal Diffusivity 10
	2.3 Flash Method 11
	2.4 Structural, Thermal and Dielectric Properties of $Ba_{1-x}Sr_xTiO_3$ and $Ca_{1-x}Sr_xTiO_3$ Ceramic Systems 14
	2.5 Summary 22
3	THEORY 24
	3.1 Flash Method 24
	3.2 Estimation of Errors and Correction 30
	3.3 Dielectric Polarization 36
	3.4 Dielectric Properties 39
	3.5 X-Ray Diffraction 41
4	METHODOLOGY 44
	4.1 Sample Preparation 44
	4.2 X-Ray Diffraction (XRD) 46
	4.3 Scanning Electron Microscope (SEM) 46
	4.4 Photoflash Technique 47
	4.4.1 The Experimental Setup 47
	a Sample and Sample Holder 48
	b Light Source 49



	c	Temperature Sensor (Thermocouple)	50
	d	The Photodiode, Preamplifier and Digital Oscilloscope	50
	4.4.2	The Experimental Procedure	51
	4.4.3	The Photoflash Technique Calibration	51
	4.5	Laser Flash Technique	51
	4.6	Impedance Spectroscopy	55
	4.6.1	Impedance Analyzer	55
	4.6.2	Electrodes and Samples	56
	4.6.3	Calibration and Experimental Procedures	57
	4.6.4	Experimental Errors in AC Impedance Analyser	57
	4.7	The Density	58
5		RESULTS AND DISCUSSIONS	59
	5.1	Introduction	59
	5.2	Structural Properties	59
	5.2.1	X-Ray Diffraction (XRD) analysis	60
	5.2.2	Surface Morphology Analysis of $Ba_{1-x}Sr_xTiO_3$ and $Ca_{1-x}Sr_xTiO_3$	71
	5.3	Thermal Diffusivity at room temperature.	76
	5.3.1	Calibration the Photoflash Technique.	76
	5.3.2	Thermal Diffusivity Measurements of Ceramics.	81
	5.3.3	The Optimum Thickness.	81
	5.3.4	Measurement of Thermal Diffusivity Values for $BaTiO_3$, $CaTiO_3$ and $SrTiO_3$.	83
	5.3.5	The Effect of Sr Substitution on Thermal Diffusivity of $Ba_{1-x}Sr_xTiO_3$ and $Ca_{1-x}Sr_xTiO_3$.	84
	5.4	Thermal Diffusivity at Medium Temperature	92
	5.4.1	Dependence of Thermal Diffusivity of $Ba_{1-x}Sr_xTiO_3$ on Temperature	92
	5.4.2	Dependence of Thermal Diffusivity of $Ca_{1-x}Sr_xTiO_3$ on Temperature	96
	5.5	Dielectric Properties	98
	5.5.1	Dependence of Dielectric Permittivity on Frequency	98
	5.5.2	The Effects of Substitution on Dielectric Constant.	101
	5.5.3	The Effects of Temperature on Dielectric Constant.	103
	5.6	Similarity of Behavior of Thermal Diffusivity and Dielectric Constant for $Ba_{1-x}Sr_xTiO_3$ and $Ca_{1-x}Sr_xTiO_3$ Ceramics	107
6		CONCLUSION	109
	6.1	Conclusion	109
	6.2	Suggestions for Future work	111

REFERENCES	112
APPENDICES	118
BIODATA OF THE STUDENT	123



LIST OF TABLES

Table		Page
3.1	Value of K_x for Various Percent Rise	30
3.2	Finite-Pulse Time Factors	34
4.1	Chemical List	44
5.1	Lattice Parameters Obtained from the Structural Refinement Using X-ray Powder Diffraction Data at Room Temperature for $Ba_{1-x}Sr_xTiO_3$.	62
5.2	Lattice Parameters Obtained from the Structural Refinement Using X-ray Powder Diffraction Data at Room Temperature for $Ca_{1-x}Sr_xTiO_3$.	69
5.3	Half Rise Time, $t_{0.5}$, Characteristic Rise Time and τ/t_c Ratio of Deferent Thickness Aluminum Samples.	78
5.4	Uncorrected Thermal Diffusivity, Correction Factor and the Corrected Thermal Diffusivity Values of Different Thickness Aluminum Samples.	79
5.5	Comparison of Thermal Diffusivity Value of Aluminum Sample with Reference Values.	80
5.6	Values of τ/t_c and the Corrected Thermal Diffusivity Values for $CaTiO_3$, $BaTiO_3$ and $SrTiO_3$ Samples.	84
5.7	Sample Thickness, Density, τ/t_c Ratio and the Corrected Thermal Diffusivity Values of BST Samples.	86
5.8	Value of τ/t_c and the Corrected Thermal Diffusivity Values of $Ca_{1-x}Sr_xTiO_3$ Samples.	91
5.9	Thermal Diffusivity Values of $Ba_{1-x}Sr_xTiO_3$ Samples at Temperature from 25 °C to 150 °C.	95
5.10	Thermal Diffusivity Values of $Ca_{1-x}Sr_xTiO_3$ Samples at Temperature from 25 °C to 150 °C.	98

LIST OF FIGURES

Figure		Page
2.1	Barium titanate perovskites structure.	14
3.1	Dimensionless plot of the rear surface temperature history.	28
3.2	Sawtooth pulse shape.	33
3.3	Parallel plate capacitor.	39
3.4	Bragg's law diffraction.	41
4.1	Experimental setup of photoflash technique.	48
4.2	Pulse shape for Minolta 5400HS photoflash.	49
4.3	Measurement part of the NETZSCH LFA 457 MicroFlash.	53
4.4	Schematic design of the sample carrier system (side view and top view) of the LFA 457 MicroFlash.	53
4.4	Schematic design of impedance analyzer	56
5.1	XRD pattern of $Ba_{1-x}Sr_xTiO_3$ ($0 \leq x \leq 1$) powder sintered at 1200 °C for 24h.	61
5.2	XRD pattern of $Ba_{1-x}Sr_xTiO_3$ ($0 \leq x \leq 0.3$) powder sintered at 1200 °C for 24h at ($44 \leq \theta \leq 48$).	64
5.3	The lattice parameters and the lattice volume as a function of x value in $Ba_{1-x}Sr_xTiO_3$.	65
5.4	XRD pattern of $Ca_{1-x}Sr_xTiO_3$ ($0 \leq x \leq 1$) powder sintered at 1200°C for 24h.	66
5.5	XRD pattern of $Ca_{1-x}Sr_xTiO_3$ ($0 \leq x \leq 0.3$) powder sintered at 1200°C for 24h ($32.5 \leq \theta \leq 33.5$).	67
5.6	XRD pattern of $Ca_{1-x}Sr_xTiO_3$ ($0.7 \leq x \leq 1$) powder sintered at 1200°C for 24h($46 \leq \theta \leq 48$).	68
5.7	The lattice parameters and the lattice volume as a function of x value in $Ca_{1-x}Sr_xTiO_3$.	70
5.8	The Micrographs of $Ba_{1-x}Sr_xTiO_3$ prepared by solid-state reaction method and sintered at 1200 °C for 24 hours: a. x=0, b. x=0.3.	72
5.9	The Micrographs of $Ba_{1-x}Sr_xTiO_3$ prepared by solid-state reaction	73



	method and sintered at 1200 °C for 24 hours; a. $x=0.5$, b. $x=0$.	
5.10	The Micrographs of $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$ prepared by solid-state reaction method and sintered at 1200 °C for 24 hours; a. $x=0$, b. $x=0.2$.	74
5.11	The Micrographs of $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$ prepared by solid-state reaction method and sintered at 1200 °C for 24 hours; a. $x=0.7$, b. $x=1$.	75
5.12	Thermogram of aluminum (thickness, $L= 0.366$ cm).	76
5.13	Thermal diffusivity for aluminum (thickness, $L=0.366\text{cm}$) calculated at various temperature rises.	77
5.14	Thermal diffusivity of aluminum (as a function of thickness).	80
5.15	Thermogram for BaTiO_3 (thickness, $L = 0.25\text{cm}$).	82
5.16	Thermal diffusivity of BaTiO_3 as a function of thickness.	82
5.17	The thermogram of CaTiO_3 , BaTiO_3 and SrTiO_3 ceramics respectively.	83
5.18	The thermal diffusivity and density as a function of x value in $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$.	85
5.19	The thermal diffusivity as a function of reciprocal density.	87
5.20	The thermal diffusivity values as a function of crystallite size [\AA].	88
5.21	The thermal diffusivity and experimental density as a function of x value in $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$.	90
5.22	Dependence of thermal diffusivity on reciprocal density of $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$.	91
5.23	Dependence of thermal diffusivity of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ ($0 \leq x \leq 0.4$) on temperature.	93
5.24	Dependence of thermal diffusivity of $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ ($0.5 \leq x \leq 1$) on temperature.	94
5.25	The dependence of thermal diffusivity of $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$ ($0 \leq x \leq 0.3$) on temperature.	96
5.26	The dependence of thermal diffusivity of $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$ ($0.4 \leq x \leq 1$) on temperature.	97
5.27	The dependent of dielectric constant for samples BaTiO_3 , SrTiO_3 and CaTiO_3 on frequency.	100



5.28	The dielectric constant values as a function of x concentration value in $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$.	101
5.29	The dielectric constant as a function of x value in $\text{Ca}_{1-x}\text{Sr}_x\text{TiO}_3$.	102
5.30	The dielectric constant versus temperature for BaTiO_3 .	104
5.31	The dielectric constant versus temperature for $\text{Ba}_{0.9}\text{Sr}_{0.1}\text{TiO}_3$.	106



LIST OF SYMBOLS

ΔL	Change in length.
ΔT	Change in temperature.
λ	Thermal conductivity
β	Adjustable parameter of the energy pulse
ω	Angular frequency
ρ	Density
τ	Pulse time
Δ	Sampling rate
α	Thermal diffusivity
α_c	Corrected value of thermal diffusivity
σ_0	DC contribution.
$\partial T/\partial x$	Temperature gradient
α_x	Thermal diffusivity calculated at x percent rise
A	Surface area
\AA	Angstrom unit
C	Capacitance
C_0	Capacitance of the empty system
c_p	Specific heat capacity
r	Distance between two conducting surfaces
D	Dielectric induction
E	Electric field
$E(\omega)$	Fourier transforms of the time dependent field

f	Frequency
G	Conductance
g	Small depth at the front surface
J_u	Heat flux
K_R	Correction factor for Taylor rising curve data
K_x	Constant corresponding to $x\%$ of maximum temperature rise
l	The mean-free path
L	Sample length
L	Sample thickness
P	Polarization
$P(\omega)$	Fourier transforms of the time dependent polarization
Q	Charge accumulated on a surface
Q	Energy of the light pulse
T	Temperature
t	Time
$t_{0.25}$	Time required to reach 25% maximum temperature rise
$t_{0.75}$	Time required to reach 75% maximum temperature rise
$t_{1/2}, t_{0.5}$	Time required for the back surface of the sample to reach half the maximum temperature rise
t_a	Time axis intercept of the temperature versus time curve
T_C	Phase transformation temperature (Curie temperature)
t_c	Characteristic rise time
T_M, T_{\max}	Maximum temperature rise at the sample rear surface
T_0	Initial temperature at the rear surface of the sample
t_x	Time required to reach $x\%$ of T_{\max}
v	Lattice vibration velocity,

V	Applied voltage
$V(\omega)$	Measured voltage
ϵ	Dielectric constant
$\epsilon(\omega)$	Dielectric constant as a function of angular frequency
ϵ^*	Complex dielectric constant as measured by the instrument
ϵ'	Real dielectric constant
ϵ_0	Dielectric constant of free space
ϵ_r	Relative dielectric constant
χ	Electric susceptibility
χ'	Real susceptibility
χ''	Imaginary susceptibility
x	Concentration of Sr^{2+} ions

LIST OF ABBREVIATIONS

SEM	Scanning Electron Microscope
XRD	X-ray Diffraction
Hz	Hertz
AC	Alternating Current
IR	Infra Red
DRAM _S	Dynamic random access memories
MLCC _S	Multi-Layer Ceramic Capacitors

CHAPTER 1

INTRODUCTION

1.1 Introduction to the Ceramics and its Applications

The term "ceramic" referred to clay-based materials. However, new generations of ceramic materials have expanded the scope and number of possible applications. Ceramic materials are inorganic compounds, usually oxides, nitrides, or carbides. The bonding is very strong either ionic or network covalent. Many adopt crystalline structures, but some form glasses. The properties of the materials are a result of the bonding and structure.

The most important thermal properties of ceramic materials are heat capacity, thermal diffusivity, and thermal conductivity. Many applications of ceramics, such as their use as insulating materials, are related to these properties. Ceramics can withstand high temperature, are good thermal insulators and do not expand greatly when heated. This makes them excellent thermal barriers, for applications such as lining industrial furnaces, thermal paint and covering the space shuttle to conserve it from high temperatures. The last generation of gas turbines hot path components (typically combustion chamber, transition pieces, rotating blades and vanes) are protected against the hot gases ($>1300^{\circ}\text{C}$) by a ceramic thermal barrier coating (TBC) with a thickness ranging from $300\ \mu\text{m}$ up to 1 or more millimeters (Cernuschi et al., 2004).



Ceramics are strong, hard, and durable and have low densities and high melting points. This makes them attractive structural materials. One significant drawback is their brittleness, but this problem was overcome by the development of new materials as example composites.

Ceramics vary in electrical properties from excellent insulators to superconductors. Thus, they are used in a wide range of applications. Some are capacitors or semiconductors in electronic devices. For example, piezoelectric materials can convert mechanical pressure into an electrical signal and are especially useful for sensors.

In recent years, much attention was devoted to the development of dielectric materials for voltage controlled, frequency-fast phase shifters and filters operating. The development in electronic and related industries on dielectric materials has created the interest to synthesize new materials with suitable properties for industrial requirements (Zhaow, 2006).

One of the electronics which attracted tremendous research interest is BaTiO_3 (barium titanate) this material has two crystallographic site, i.e. A and B site and the modification can lead to new materials with different properties (Walter, 2005). Ferroelectrics such as $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ (barium strontium titanate) have emerged as leading candidates for electronic applications due to their highly nonlinear dielectric