UNIVERSITI PUTRA MALAYSIA

PARALLEL EVOLUTION OF MICROSTRUCTURE, THERMOPHYSICAL AND ELECTRICAL PROPERTIES OF UNDOPED AND LA-DOPED STRONTIUM TITANATE

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ITMA 2016 19

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By

IDZA RIATI BINTI IBRAHIM

Thesis submitted to the School of Graduate Studies, Universiti Putra Malaysia, in fulfilment of the Requirement for the Degree of Doctor of Philosophy

November 2016

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Doctor of Philosophy

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November 2016

Chairman: Assoc. Prof. Khamirul Amin bin Matori, PhD
Institute: Institute of Advanced Technology

Thermophysical and electrical properties of undoped and lanthanum-doped strontium titanate have been largely focusing on the final product of the final sintering temperature, thus neglecting the parallel evolution of microstructure and material properties at various intermediate sintering temperatures. Therefore, much possible essential information has been neglected, thus reducing the capabilities of producing good fundamental scientific knowledge of the microstructure-material properties, particularly in thermophysical and electrical properties. In order to filling up the vacuum, a systematic development study from nanometric to micronic grains with parallel evolution of thermophysical and electrical properties has been elucidated. Strontium titanate, undoped and lanthanum-doped, have been prepared via the high energy ball milling and followed by sintering from 500 to 1400 °C for different microstructural features. The XRD patterns showed an improvement of crystallinity from amorphous + crystalline mixture phases to a complete polycrystalline phase with increasing sintering temperature. The Raman spectra showed the presence of second-order features for fully-formed phase. ESR spectra revealed the paramagnetic defects existed in undoped and lanthanum-doped strontium titanate. FESEM micrographs showed the grains evolved from 38.3 nm to 2.6 µm for without presintered strontium titanate and 38.0 nm to 9.1 µm for presintered strontium titanate. The grains size fluctuated, attributing to carbonate decomposition but became larger as the sintering temperature increased from 800 to 1400 °C due to the grain growth phenomenon. Doping of lanthanum has tendency to inhibit the grain growth where significant reduction of grain size (range from 34 nm to 0.47 µm) could be observed. Without the presence of other leading factors such as porosity, thermal diffusivity and conductivity increased with increased grain size with the maximum value of 2.97 mm^2/s and 7.09 W/m.K, respectively, exhibited by the presintered strontium titanate sample. The electrical conductivity was improved with increased grain size due to reduced amount of grain boundary resistivity. It can be concluded that the phonon scattering mechanisms and electrical transport properties

were intrinsically and extrinsically influenced by several factors. Intrinsic contribution comes from the crystal structure evolving from highly amorphous to a complete polycrystalline strontium titanate. Extrinsic contributions by the microstructure, particularly from the impurities, grain boundaries, grain size, density and the porosity influenced the phonon-impurities, phonon-defects and phonon-boundaries scattering, so also the barrier to the flow of electrons.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsafah

EVOLUSI SELARI SIFAT-SIFAT MIKROSTRUKTUR, TERMOFIZIKAL, DAN ELEKTRIKAL UNTUK STRONTIUM TITANAT TANPA DOP DAN DIDOPKAN LANTANUM

Oleh

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November 2016

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Sifat-sifat termofizikal dan elektrik strontium titanat tanpa dop dan strontium titanat didopkan lantanum sangat menumpukan kepada produk akhir bagi suhu pensinteran akhir, lalu mengabaikan evolusi selari sifat-sifat mikrostruktur dan sifat-sifat bahan pada pelbagai suhu pensinteran di perantaraan. Maka, banyak maklumat penting mungkin telah diabaikan, dengan itu mengurangkan keupayaan penghasilan pengetahuan saintifik asas yang baik yang terletak di sebalik evolusi selari sifat mikrostruktur-bahan, khususnya dalam sifat-sifat termofizikal dan elektrik. Untuk mengisi ruang vakum tersebut, suatu kajian perkembangan sistematik dari butiran bersaiz nano ke mikron dengan evolusi selari sifat-sifat termofizikal dan elektrikal telah dijelaskan. Strontium titanat, tanpa dop dan didop dengan lantanum, telah disediakan melalui pengisaran bebola bertenaga tinggi dan seterusnya pensinteran dari suhu 500 ke 1400 °C untuk menghasilkan sifat mikrostruktur yang pelbagai. Corak-corak XRD menunjukkan peningkatan penghabluran dengan peningkatan suhu pensinteran dari fasa campuran amorfus + kristal ke fasa polihabluran lengkap. Spektrum Raman menunjukkan kehadiran tertib kedua untuk fasa yang telah terbentuk sepenuhnya. Spektrum ESR mendedahkan kecacatan paramagnet wujud dalam strontium titanat tanpa dop dan yang didop lanthanum. Mikrograf FESEM menunjukkan butiran berevolusi dari 38.3 nm ke 2.6 µm untuk strontium titanat tanpa pra pensinteran dan and 38.0 nm ke 9.1 µm untuk strontium titanat dengan pensinteran. Saiz butiran turun naik disebabkan oleh penguraian karbonat tetapi meningkat menjadi lebih besar kerana suhu pensinteran meningkat dari 800-1400 °C disebabkan fenomena pertumbuhan butiran. Pengedopan lantanum ke dalam strontium titanat telah mempunyai kecenderungan untuk menghalang pertumbuhan butiran di mana pengurangan ketara saiz butiran (berjulat di antara 34.0 nm ke 0.5 µm) dapat diperhatikan. Tanpa kehadiran faktor-faktor utama yang lain seperti keliangan, kemeresapan terma dan kekonduksian meningkat dengan peningkatan saiz butiran dengan nilai maksimum 2.97 mm^2/s dan 7.09 W/m.K masing-masing yang dipamerkan oleh sampel strontium titanat dengan pra pensinteran. Kekonduksian

elektrik telah bertambah baik dengan peningkatan saiz butiran kerana pengurangan kerintangan sempadan butiran. Dapat disimpulkan bahawa mekanisma penyerakan fonon dan sifat-sifat pengangkutan elektrik adalah dipengaruhi oleh beberapa faktor ekstrinsik dan intrinsik. Sumbangan intrinsik datang daripada struktur hablur yang berevolusi dari sangat amorfus ke polihabluran lengkap strontium titanat. Sumbangan ekstrinsik oleh mikrostruktur, terutamanya bendasing, sempadan butiran, saiz butiran, ketumpatan dan keliangan mempengaruhi penyerakan fonon-bendasing, fonon-kecacatan dan fonon-sempadan, begitu juga halangan kepada pergerakan elektron-elektron.

ACKNOWLEDGEMENTS

First praise is to Allah, the Almighty, on whom ultimately we depend for sustenance and guidance. With Allah blessing, help and will, we were able to make this work a success.

My sincere appreciation and gratitude goes to my former supervisor Allahyarham Dr. Mansor Hashim who initiated this evolution works on 2008 and has been showed his tremendously continuous guidance, advice and support since then until the end of his life. The evolution works which initially covered on magnetic materials and magnetic properties has been currently expanded into other nanostructured polycrystalline materials. With his timely and efficient contribution during his life helped me shape this into its final form and I express my sincerest appreciation for his assistance in any way that I may have asked. May Allah grant him a place in Jannatul Firdaus and be placed amongst the blessed alongside Allah S.W.T.

I would also like to extent my sincerest thanks and appreciation to my current supervisor, Assoc. Prof. Dr. Khamirul Amin Matori for his time, effort, helps and guidance in ensuring the successful completion of this project and thesis writing. I am also very much thankful to my other supervisory committee, Assoc. Prof. Dr. Zaidan Abd. Wahab and Dr. Ismayadi Ismail for their valuable guidance, constructive criticism and encouragement throughout this project. I would also like to express my gratitude to Prof. Madya Dr. Jumiah Hassan, Prof. Madya Dr. Raba'ah Syahidah Azis, Dr. Norlaily Mohd Saiden and Dr. Nurhidayaty Mokhtar for their great helps and contributions throughout the research project.

I am highly grateful for the love, care, prayers and support from the love of my life, my mother, Mrs. Razilah Othman and my brother, Mr. Idzharudin Ibrahim who have made the accomplishment of this project possible. Their strong support was the keen motivation for me to keep moving forward in this research journey. To my late father, Allahyarham Ibrahim Nor, my thoughts and prayers are always be with you. May Allah S.W.T. bless you with Jannatul Firdaus and be placed amongst the blessed alongside Allah S.W.T.

To my colleagues in Nanostructured Polycrystals and Magnetism Group (NanoMag), Dr. Ismayadi Ismail, Dr. Samikannu Kanagesan, Dr. Shamsul Ezzad, Dr. Abdollah Hajalilou, Dr. Rahim Sabbaghizadeh, Ms. Rodziah Nazlan, Ms. Nor Hapishah Abdullah, Mrs. Wan Norailiana Wan Ab Rahman, Mrs. Fadzidah Mohd Idris, Mrs. Noor Lyana Adnan, Ms. Ghazaleh Bahmanrokh, Ms. Nuraine Mariana Mohd Shahrani, Ms. Yusnita Yusuf, Ms. Eydar Tey, Ms. Anwar Ali Baqer, Mrs. Mutia Suhaibah Abdullah, Mrs. Dayang Nur Fazliana Abd Halim, Ms. Farah Nabilah Shafiee, Mrs. Azdiya Suhada Abdul Rahim, Ms. Nur Izzaiti Ibrahim, Mr. Muhammad Misbah Muhamad Zulkimi, Mr. Muhammad Syazwan Mustaffa, Mr. Low Zhi Huang, Mr. Noruzaman Daud, Mr. Muhammad Aizat Noor Ismail, and Mr. Che Sulaiman Ahmad, I appreciate the discussions, ideas, memorable interactions and time spent throughout this research. To my fellow friends during my

postgraduate study, Ms. Noorfauzana Adnin, Ms. Nur Fadilah Baharuddin, Ms. Nurzilla Mohammad, Ms. Zamratul Maisarah Mohd Ismail, and many others, I say thanks for walking together in this doctoral journey and May Allah ease your ways.

Lastly, I would also like to express my gratitude to all of Institute of Advanced Technology and Faculty of Science staff, Mr. Mohd Kadri Masaud, Mrs. Sarinawani Abdul Ghani, Mr. Ali Rani, Mrs. Rosnah Nawang, Mrs. Roslina Abdul Rashid, Ms. Nurnazeera Zukefli, Mrs. Noor Lina Shamsuddin, Mrs. Roslina Warno, Mrs. Noor Linda Hassan, Mrs. Rokiah Deraman, Mrs. Norhaslinda Noruddin, Mrs. Kamsiah Alias, Mr. Roslim Mohd, Mr. Mohd Zain Mohd Yusof and many others for their great helps and contributions.

I certify that a Thesis Examination Committee has met on 25 November 2016 to conduct the final examination of Idza Riati binti Ibrahim on her thesis entitled "Parallel Evolution of Microstructure, Thermophysical and Electrical Properties of Undoped and La-Doped Strontium Titanate" 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 Doctor of Philosophy.

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

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LIST OF ABBREVIATIONS AND SYMBOLS

$SrTiO_3$	Strontium titanate
$La_{0.08}Sr_{0.92}TiO_3$	Lanthanum-doped strontium titanate
WPS-STO	Without presintered strontium titanate
PS-STO	Presintered strontium titanate
La-STO	Lanthanum-doped strontium titanate
ZT	Thermoelectric figure of merit
vol.	Volume
wt.	Weight
T_q	Quantum paraelectric phase transition temperature
T_c	Critical temperature
α	Thermal diffusivity
k	Thermal conductivity
c	Specific heat
λ	Mean free path
DC	Direct current
ρ_{DC}	Electrical resistivity
σ	Electrical conductivity
TEM	Transmission Electron Microscopy
XRD	X-ray Diffraction
FESEM	Field Emission Electron Microscopy
TGA	Thermogravimetric Analysis
DSC	Differential Scanning Calorimetry

ESR	Electron Spin Resonance
EDX	Energy Dispersive X-ray Spectroscopy
ρ	Density
LFA	Laser Flash Apparatus
ρ_{xrd}	X-ray density
ρ_{exp}	Experimental density
k_B	Boltzmann constant

CHAPTER 1

INTRODUCTION

1.1 Background of the study

Materials' properties concerning thermophysical and electrical properties have been greatly studied by materials scientists and engineers for so many decades, then becoming sensations in recent years due to high technological demand on nanomaterials and nanotechnology. This interest in nanomaterials and nanotechnology is mainly due to their extraordinary physical and chemical properties relative to the corresponding properties present in bulk materials. Thermophysical and electrical properties of the materials are determined by the intrinsic and extrinsic properties in the materials. The intrinsic properties are influenced by the structure at the atomic scale and not susceptible to significant change by modification of the microstructure, such as the melting point, elastic modulus, coefficient of thermal expansion, and whether the material is brittle, magnetic, ferroelectric, or semiconducting. In contrast, many of the properties critical to the engineering applications of materials are strongly dependent on the microstructure such as mechanical strength, dielectric constant, and electrical conductivity (Rahaman, 2003).

Microstructure of polycrystalline ceramics is usually complex, consisting of grains, grain boundaries, porosity and secondary phases. This kind of structure is not seen in single crystals. The shape, size, distribution and orientation of the grains play a key role in many of the macroscopic properties including thermophysical and electrical properties. Grain boundaries and additional phases were thought to be undesirable, and the goal was to eliminate them and obtain a structure as close as to single crystals as possible (Ichinose et al., 1987). However, with new research and technology developments, these surfaces and grain boundaries have been utilized as positive tools to enhance certain important properties including thermophysical and electrical properties in ceramics. The variation in thermophysical and electrical properties are resulting from variation microstructural features due to presence of impurities, porosity, dislocations and various sizes of grains. The presence of impurities disrupts the normal lattice arrangement, thus decreasing the heat transfer and electrical flow. Essentially, these phenomenons are familiar with the polycrystalline ceramics samples having micrometer grain size and the information on their relationships is well understood. Materials in the micrometer scale mostly exhibit physical properties the same as that of bulk form; however materials in the nanometer scale may exhibit physical properties distinctively different from that of micrometer scale (Kinemuchi et al., 2010). Nanomaterials may have significantly lower melting point or phase transition temperature and appreciably reduced lattice constants, due to a huge fraction of surface atoms in the total amount of atoms (Cao and Wang, 2011). Materials with a modified 'nano'-microstructure provide potential for new or improved applications (Winterer, 2002).

Therefore, in this study we intend to explore the underlying science of rarely untouched parallel evolving microstructure with material properties (particularly thermophysical and electrical properties) from several nanometers up to micrometer grain size to elucidate the nature behind these parallel properties. Consequently, parallel evolution study of materials' properties with their particular microstructural properties delivers great information specifically on current missing intermediate developments of materials located at nano-grain sized regime. The example of the parallel evolution-data acquisition scheme is depicted in Figure 1.1.

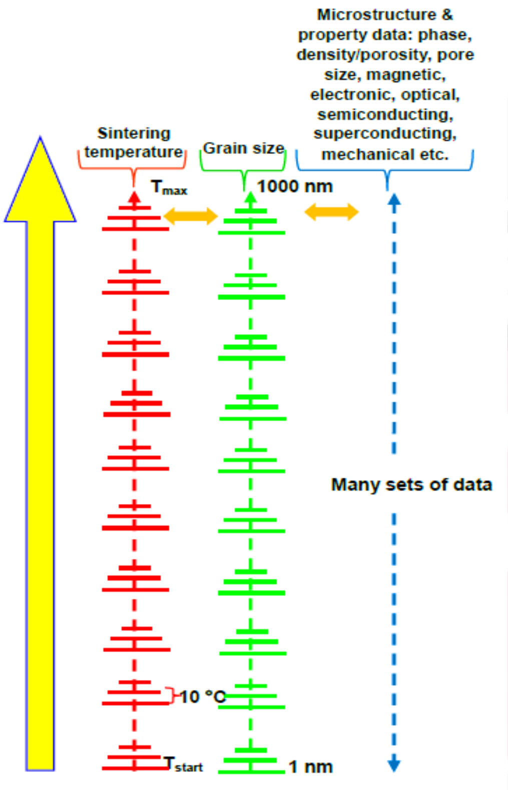

Figure 1.1: Parallel evolution-data acquisition scheme

In this research, the raw constituents of strontium oxide, titanium dioxide and strontium carbonate powder for preparation of undoped strontium titanate and addition of small amount of lanthanum oxide powder for preparation of lanthanum-doped strontium titanate were milled using high energy ball milling. This milling process is carried out in order to produce nanometer starting powder with significant amount of amorphous phase. Later, sintering would be performed onto the bulk samples from unusually low sintering temperature (500 °C) to somewhat high sintering temperature (1400 °C) to produce microstructural variations in the samples. Afterward, the parallel evolution of microstructure-material properties particularly thermophysical and electrical properties were elucidated.

1.2 Selection of materials

There are major applications that utilize the varied thermophysical and electrical properties of ceramics. The applications could be ranging from high thermal conductivity substrates for electronic packaging to low thermal conductivity with high electrical conductivity in thermoelectric materials. It has been evidenced that strontium titanate, $SrTiO_3$, is a promising candidate for applications in thermoelectric, thermal management applications and modern electronic devices. It is highly attractive because of its desirable thermal, chemical stability and semiconducting behaviour. $SrTiO_3$ is a well-known quantum paraelectric material and has been greatly garnered interest both for experimentalists and theorists since 1950 (Hulm, 1950).

Various studies had shown that by introducing oxide defects or doped with alkaline earth metals or transition metals in $SrTiO_3$, its electrical, thermal and thermoelectric properties can be modified. It has been reported that lanthanum-doped $SrTiO_3$ increases electrical conductivity and suppresses the heat transport (Koumoto et al., 2010; Shang et al., 2010a). This compound is still open and still attracts much attention since it can be regarded as a model system for perovskite ceramics due to its stability in the cubic crystal system above –168 °C (Syha, 2014) and model substance that is still closely related to the industrially used modified barium or lead titanates (Moos and Hardtl, 1997). Accordingly, $SrTiO_3$ is chosen as a suitable material for evolution study particularly in their thermophysical and electrical properties for these reasons.

1.3 Problem statement

Microstructural dependence of thermophysical and electrical properties for polycrystalline $SrTiO_3$ having micrometer grain size has been widely studied and greatly understood. However, research on $SrTiO_3$ with the nanometer scale has been a field of intense study, due to the novel properties shown by particles located in the transition region between the isolated atoms and bulk solids. Their novel properties make them attractive, both from the scientific knowledge of understanding their properties, and the technological importance of enhancing the performance of the present materials. Extensive research had been merely carried out on sample at final sintering temperature by neglecting the parallel evolution of microstructure and material properties at various intermediate sintering temperatures. Therefore, much possible essential development information has been neglected, thus reducing the capabilities of producing good fundamental scientific knowledge which lies behind the parallel evolution of the microstructure-material properties, particularly in thermophysical and electrical properties.

This absence information has leaving behind many research questions that have to be solved in this study:

i. How microstructural properties evolve with the thermophysical and the electrical properties from several nanometers to micrometer grains size?
ii. How the evolution of an amorphous-crystalline mixture state to complete polycrystalline state affects the microstructure-material properties?

iii. Does sample with nanometer grain size demonstrates the similar material properties with samples having micron grain size?
iv. What is the relationship of evolving microstructure properties with thermophysical and electrical properties of material?
v. When is the remarkable transition of material properties between the nanometer and micrometer grain size?

Hence, through this exploratory research project, the study intends to solve and reveal the systematic development of grains having size from several nanometers up to micrometer with evolving thermophysical and electrical properties which has yet to be established.

1.4 Objectives and hypotheses of the study

The vital goal of this research is to critically track down the parallel evolution of microstructural, thermophysical and electrical properties from unusually lower sintering temperature (amorphous + crystalline mixture state) to somewhat higher temperature (complete polycrystalline state).

Hence, this study embarks on the following main objectives:

i. To investigate the microstructure-thermophysical and electrical properties parallel evolving relationships from an amorphous + crystalline mixture state to a complete polycrystalline $SrTiO_3$ and lanthanum-doped $SrTiO_3$.
ii. To study the development and analyze the physical interpretation of parallel evolving microstructure-thermophysical and electrical properties of $SrTiO_3$ lanthanum-doped $SrTiO_3$.

Thus according the above main objectives, this study is hypothesized as follows:

i. Polycrystalline $SrTiO_3$ and lanthanum-doped $SrTiO_3$ with micronic grains microstructure would result in complete crystalline phase, thus increasing the thermophysical and electrical properties. However, the existence of amorphous + crystalline mixture state with nanocrystalline microstructure would deteriorate the thermophysical and electrical properties.
ii. As the undoped and lanthanum-doped $SrTiO_3$ evolved from nanometric to micronic grains, the thermophysical properties would increase due to less phonon scattering effects in the samples, thus increasing the thermal difuusivity and thermal conductivity. It is expected that the electrical resistivity would decrease with increasing grain size due to reduce amount of grain boundaries that acting as barrier to the movement of electrons.

In order to achieve the main objectives of the study, these following work-phase objectives have been carried out which are:

i. To synthesize nanoparticles size powders of raw constituents of undoped and lanthanum-doped $SrTiO_3$ via high energy ball milling.
ii. To produce different morphological variations of undoped and lanthanum-doped $SrTiO_3$ via sintering of bulk compressed powder at different sintering temperatures.

iii. To carry out the parallel microstructural with thermophysical and electrical measurements for undoped and lanthanum-doped $SrTiO_3$.

Thus according the above work-phase objectives, the results are expected as follows:

i. Nanoparticles size starting powder from raw constituents of undoped and lanthanum-doped $SrTiO_3$ would be synthesized after 12 hours high energy ball milling.
ii. Grain size of undoped and lanthanum-doped $SrTiO_3$ would increase with increase in sintering temperature.
iii. The data of microstructural measurement would show a strong relationship with thermophysical and electrical measurement where the thermophysical and electrical data would enhance with the improvement in microstructural formation of undoped and lanthanum-doped $SrTiO_3$.

1.5 Outline of the thesis

This study is aimed to elucidate parallel evolution of microstructure, thermophysical and electrical properties and their relationships towards a complete polycrystalline $SrTiO_3$. It begins with the introductory part in Chapter 1. Background of study, problem statement and objectives of the study are described in this chapter. Chapter 2 focuses on recent progress in relevant field of study, synthesis method of nannocrystalline $SrTiO_3$, reviews on physical and microstructural properties, thermophysical properties and activation energy of electrical conductivity of $SrTiO_3$-based materials, some influence of microstructural properties on thermal and electrical properties. Research theories are briefed and presented in Chapter 3. Theories on the thermophysical and electrical properties will be exposed in this chapter. The thesis outline is continued with research methodology in Chapter 4. In this chapter, research design is discussed in terms of materials preparation and characterization. Next, the obtained results from this study would be discussed in Chapter 5. Detailed explanations and summary of the obtained trends will be presented in this chapter. Finally, the conclusions and achievements of the study will be concluded in the final chapter of the thesis (Chapter 6).

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