UNIVERSITI PUTRA MALAYSIA

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MASTER OF SCIENCE
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By

SITI NOR AIN BINTI RUSLY

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

February 2013

DEDICATION

I dedicate this thesis to my beloved husband, parent, family and friends.

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

MICROSTRUCTURAL, MAGNETIC AND DIELECTRIC PROPERTIES OF $Bi_{1-x}Sm_xFeO_3$ MULTIFERROIC MATERIALS

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SITI NOR AIN BINTI RUSLY
February 2013

Chairman: Professor Abdul Halim Bin Shaari, PhD

Faculty: Science

$BiFeO_3$ (BFO) is a most common type of multiferroic materials that exhibits antiferromagnetic and ferroelectric order at room temperature. Based on previous reports, it was rather difficult to synthesize BFO in form of pure single phase due to narrow range of temperature stabilities. Hence, in this thesis, we report some research findings on the effect of different small ranges of calcinations and sintering temperature for preparing BFO. The best BFO sample could be determine by analyzing the phase transformation, magnetic and dielectric properties using XRD, VSM and impedance analyzer respectively. The effect of Sm substitution in the BFO system also has been studied. Samples of $Bi_{1-x}Sm_xFeO_3$ (BSFO) with x = 0.0, 0.1, 0.2, 0.3, 0.4 and 0.5 were prepared using solid state reaction method. There are three series of samples that have been prepared which are Sample A (both calcinations and sintering temperature at 800 ºC), Sample B (calcinations temperature at 800 ºC and sintering temperature at 825 ºC) and Sample C (both calcinations and sintering

temperature at 825 ºC). The XRD pattern showed an improvement of crystallinity in pure BFO with the lower unwanted secondary phases by increasing the calcinations and sintering temperature at 825 ºC. However, the unwanted secondary phases disappeared in BSFO sample implying that Sm^{3+} substitution can stabilize the perovskite structure. SEM micrograph showed a well defines grain structures with clear grain boundaries in BFO sample. A larger grain sizes were observed as the calcinations and sintering temperature increase. However, BSFO have smaller average grain size than BFO sample. As the Sm composition increases from x = 0.0 to x = 0.5, the density values decreased for all series. The density increases proportionally with sintering temperature caused by elimination pores. The magnetization analysis showed that BFO compound have very narrow hysteresis loop exhibits antiferromagnetic behavior (Hc = 191 Oe and Mr = 1.81 x 10^{-3}emu/g) at room temperature. The result showed magnetic properties were enhanced with higher calcinations and sintering temperature at 825 ºC. Larger hysteresis loop were obtained in BSFO indicates weak ferromagnetic behavior and the magnetization values increases when Sm composition increases. Sample C5 have highest magnetic properties with Hc = 3589.9 Oe and Mr = 7.52 x 10^{-2} emu/g. The dielectric permittivity, ε′ and dielectric loss, ε″ decreased with increasing of frequency. The higher calcinations and sintering temperature, Sample C has the higher value of ε′ and ε″. The value of ε′ and ε″ increased with Sm composition and dielectric measuring temperature. The dispersion of ε′ and ε″ are maximum for Sample C5 with ε′ ~ 141 and ε″ ~ 5 at room temperature. Hence BSFO with x =0.5 with higher calcinations and sintering (825 ºC) is formed to be a better multiferroic material than pure BFO sample by resulting enhancement in magnetic and dielectric properties.

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

MIKROSTRUKTUR, SIFAT MAGNET DAN DIELEKTRIK BAGI BAHAN MULTIFEROIK $Bi_{1-x}Sm_xFeO_3$

Oleh

SITI NOR AIN BINTI RUSLY
Februari 2013

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$BiFeO_3$ (BFO) merupakan salah satu bahan multiferoik yang menunjukkan sifat antiferomagnet dan feroelectrik pada suhu bilik. Kajian lepas menunjukkan agak sukar untuk mensintesis BFO dalam bentuk fasa tulen tunggal disebabkan oleh julat suhu kestabilan yang kecil. Maka, dalam tesis ini kami melaporkan beberapa penemuan penyelidikan tentang kesan perbezaan julat kecil suhu pengkalsinan dan pensinteran bagi penyediaan BFO. Sampel BFO yang terbaik dapat ditentukan dengan menganalisis pembentukan fasa, sifat magnet dan sifat dielektrik masing-masing dengan menggunakan XRD, VSM dan penganalisis impedans analyzer. Kesan penggantian Sm dalam sistem BFO juga telah dikaji. Sampel $Bi_{1-x}Sm_xFeO_3$ (BSFO) dengan x = 0.0, 0.1, 0.2, 0.3, 0.4 and 0.5 telah disediakan menggunakan kaedah tindakbalas keadaan pepejal. Tiga siri sampel telah disediakan dalam kajian ini, iaitu Sampel A (Kedua-dua suhu pengkalsinan dan pesinteran pada 800 ºC), Sampel B (Suhu pengkalsinan pada 800 ºC dan suhu pesinteran pada 825 ºC) dan Sampel C (Kedua-dua suhu pengkalsinan dan pesinteran pada suhu 825 ºC). Corak

pembelauan sinar-X menunjukkan penambahbaikan penghabluran dalam BFO dengan kandungan fasa sekunder tidak dikehendaki yang rendah dengan kenaikan suhu pengkalsinan dan pensinteran pada suhu 825 ºC. Walaubagaimanapun, fasa sekunder tidak dikehendaki tidak dikesan dalam sampel BSFO memperihalkan bahawa penggantian ion Sm dapat menstabilkan struktur perovskit. Mikrograf SEM telah menunjukkan struktur butiran sempurna dengan sempadan butiran yang jelas bagi sampel BFO. Saiz butiran yang lebih besar diperolehi apabila suhu pengkalsinan dan pesinteran meningkat. Walaubagaimanapun, BSFO mempunyai saiz purata butiran yang lebih kecil daripada BFO. Dengan bertambahnya kandungan Sm daripada x = 0.0 kepada x = 0.5, nilai ketumpatan bahan berkurangan bagi semua siri. Nilai ketumpatan meningkat berkadaran dengan suhu pensinteran disebabkan oleh penyingkiran liang-liang. Analisis kemagnetan menunjukkan bahawa sebatian BFO mempunyai gelung histeresis yang sempit mempamerkan sifat antiferomagnetik (Hc = 191 Oe and Mr = 1.81 x 10^{-3} emu/g) pada suhu bilik. Hasil kajian menunjukkan sifat magnet meningkat dengan peningkatan suhu pengkalsinan dan suhu pesinteran pada 825 ºC. Gelung histeresis yang lebih besar diperolehi dalam BSFO menunjukkan sifat feromagnetik lemah dan nilai kemagnetan bertambah apabila komposisi Sm bertambah. Sampel C5 mempunyai sifat magnet yang tertinggi dengan Hc = 3589.9 Oe dan Mr = 7.52 x 10^{-2} emu/g. Nilai ketelusan dielektrik, ε′ dan dielektrik lesapan, ε″ meningkat dengan kenaikan frekuensi. Sampel BSFO yang dikalsin dan disinter pada suhu tinggi 825 ºC mempunyai nilai ε′ dan ε″ yang tinggi. Nilai ε′ dan nilai ε″ bertambah dengan kenaikan komposisi Sm dan suhu pengukuran dielektrik. Penyebaran ε′ dan ε″ adalah maksimum untuk Sampel C5 dengan nilai ε′ ~ 141 dan ε″ ~ 5 pada suhu bilik. Maka, BSFO dengan x = 0.5 dengan pengkalsinan dan pesinteran pada suhu tertinggi (825 ºC) merupakan bahan multiferroik yang lebih

baik daripada sampel asli BFO dengan penambahbaikan sifat magnet dan sifat dielekrik.

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.

I certify that a Thesis Examination Committee has met on **1 FEBRUARI 2013** to conduct the final examination of **SITI NOR AIN BINTI RUSLY** on her Master thesis entitled **"MICROSTRUCTURAL, MAGNETIC AND DIELECTRIC PROPERTIES OF $Bi_{1-x}Sm_xFeO_3$ MULTIFERROIC MATERIALS"** 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 relevant degree.

Members of the Thesis Examination Committee were as follows:

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This thesis 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 Supervisor Committee were as follows:

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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.

SITI NOR AIN BINTI RUSLY

Date: 1 February 2013

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