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DIELECTRIC AND MAGNETIC CHARACTERIZATION OF (La0.5-xPrxBa0.5)(Mn0.5Ti0.5)O3 PEROVSKITE AS A MULTIFERROIC MATERIAL SYNTHESIZED VIA SOLID STATE TECHNIQUE

NOR HAYATI BT. ALIAS

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By

NOR HAYATI BT. ALIAS

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

October 2009

DEDICATION

To god, my children and Dear husband

For the remembrance of my beloved late mother and father,

Friends, Universiti Putra Malaysia and Agency Nuclear Malaysia

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

DIELECTRIC AND MAGNETIC CHARACTERIZATION OF $(La_{0.5-x}Pr_xBa_{0.5})(Mn_{0.5}Ti_{0.5})O_3$ PEROVSKITE AS A MULTIFERROIC MATERIAL SYNTHESIZED VIA SOLID STATE TECHNIQUE

By

NOR HAYATI BT. ALIAS

October 2009

Chairman: Abdul Halim Shaari, PhD.

Faculty: Science

A new manganate perovskite $(La_{0.5-x}Pr_xBa_{0.5})(Mn_{0.5}Ti_{0.5})O_3$ has been prepared by ceramic solid-state technique at sintering temperature 1300 °C for 24 hours. The x concentration of Praseodymiun (Pr) in molar proportion in A site has been varied as x = 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, 007, 0.08, 0.09, 0.1, 0.2, 0.3 and 0.4. The dielectric properties of the synthesized materials have been studied at frequency ranging from 5 Hz to 1 MHz from room temperature up to 200 ˚C. Furthermore, analyses have been carried out to determine the structural, magnetic and dielectric electrical properties of the synthesized material as a candidate of multiferroic material.

Pr (Praseodymium) addition would promote liquid phase sintering in the synthesized samples. This enhanced the agglomeration and porosity formation in the bulk volume. The defects and conducting liquid present would generate traps that

produce negative permittivity response at interfacial/space charge frequency region due to delay in transient current or situation in which inertial conducting current of the trap presents, exceeding the charging-discharging current component. The XRD (X-Ray Diffractometry) results indicate all samples possess a single phase monoclinic structure with space group P112. Multiferroic magnetodielectric coupling in material with ferromagnetic and high dielectric constant (> 30) is able to achieve in sampel with x molar concentration = 0.07 at room temperature. The dielectric value obtained is 176 with loss tan δ value 0.62. SEM/EDX analysis of the sample shows fine grain microstructure and high manganese (Mn) content (> 0.6 wt %) which favours the double exchange mechanism.

Comparing to unsubstituted Pr sample, x=0; sample with x molar concentration 0.2 and 0.4 shows enhanced dielectric values with additional loss. The ranged of dielectric value obtained for unsubstituted sample x=0 is 3117 to 12396 while for x=0.2 and x=0.4 the value range obtained is 1611 to 16316 and 967 to 13185 respectively. The increment is associated with both polarization and conduction mechanism process. Where, the dielectric constant and loss is contribution from the effect of space charge polarization and/or ion conducting motion. Respectively, generally higher values of dielectric constant is obtained in unsubstituted sample x = 0 and high x substituted concentration 0.1, 0.2, 0.3 and 0.4 as a results of dual relaxational polarization mechanisms existed in the frequency ranged studied. Dc conduction is found to be dominated the sample at high temperature regime and for low substituted sample with x molar concentration 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08 and 0.09. This is to suggest of enhanced mobility of localized charge carriers in liquid phase region. Whereas in sample Pr(X)0.07, dominated dc conduction effect is due to fast ion hopping conduction in its fine grain microstructure with high Mn content.

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

PENCIRIAN DIELEKTRIK DAN MAGNET $(La_{0.5-x}Pr_xBa_{0.5})(Mn_{0.5}Ti_{0.5})O_3$ PEROVSKITE SEBAGAI BAHAN MULTIFERROIC MELALUI TEKNIK KEADAAN PEPEJAL

Oleh

NOR HAYATI BT. ALIAS

Oktober 2009

Pengerusi: Abdul Halim Shaari, PhD.

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Satu bahan baru seramik perovskite dari kumpulan manganit $(La_{0.5-x}Pr_xBa_{0.5})(Mn_{0.5}Ti_{0.5})O_3$ telah disediakan melalui teknik tindak balas keadaan pepejal pada suhu pensinteran 1300 ° C selama 24 jam. Kepekatan x molar Praseodymium (Pr) pada kedudukan A pada perovskite telah ditetapkan sebagai x = 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, 007, 0.08, 0.09, 0.1, 0.2, 0.3 and 0.4. Kajian dielektrik telah dikendalikan pada julat frekuensi 5 Hz sehingga 1 MHz dalam keadaan suhu bilik sehingga suhu mencapai 200˚ C. Analisa juga dilakukan untuk mengkaji sifat struktur, magnet dan dielektrik bahan yang telah disintesis serta melihat kesan pengkupelan multiferoik pada bahan yang dikaji.

Telah didapati bahawa kesan penggantian Pr (Praseodymium) ke atas bahan telah meningkatkan kesan pembentukan fasa solidus cecair pada mikrostruktur bahan.

Kesan agglomerasi dan pembentukan liang pada bahan juga adalah agak ketara. Ini memberi kesan kecacatan dalam bahan. Kecacatan serta kandungan fasa solidus cecair dalam mikrostruktur bahan telah mewujudkan kawasan negatif permittiviti spektra pada julat frekuensi interfasial. Ini adalah mungkin disebabkan oleh kesan kelambatan pada pergerakan arus peralihan elektrik atau disebabkan oleh kesan penghasilan arus inertia daripada perangkap kecacatan yang lebih besar. Analisis XRD (X-Ray Difraktometri) telah menunjukkan bahan yang disintesis mempunyai fasa tunggal hablur monoklinik P112. Adalah didapati juga bahawa kesan pengkupelan sifat dielektrik dan magnet berjaya diperolehi pada x dengan kepekatan molar 0.07. Sifatnya adalah feromagnatik dengan nilai pemalar dielektrik bahan 176 dan tan δ 0.76 pada suhu bilik. Bahan ini juga didapati mempunyai mikrostruktur dengan saiz butiran yang kecil serta kandungan Mangan (Mn) yang agak tinggi (> 0.6 bt %) lantas menambahkan kesan mekanisma pertukaran ganda dua (DE) pada bahan.

Jika dibandingkan dengan bahan tanpa penggantian Pr iaitu x=0; sampel x=0.2 dan x=0.4 menunjukkan peningkatan nilai pemalar dielektrik juga nilai tan δ nya. Julat nilai dielektrik yang diperoleh untuk sampel x=0 adalah 3117 to 12396. Sementara julat nilai yang diperoleh dari sampel dengan x molar kepekatan x=0.2 dan x=0.4 ialah 1611 - 16316 dan 967 - 13185. Keadaan ini adalah disebabkan oleh kedua-dua jenis mekanisma yang hadir pada bahan iaitu kesan relaksasi polarisasi serta kekonduksian. Dimana nilai pemalar dielektrik dan tan δ yang diperolehi adalah gabungan daripada kesan polarisasi interfasial dan/atau konduksi pergerakan ion. Nilai pemalar dielektrik dengan x molar = 0, 0.1, 0.2, 0.3 dan 0.4 juga didapati lebih tinggi berbanding sampel x molar rendah. Ini disebabkan oleh wujudnya kesan dwirelaksasi polarisasi bahan pada campuran kepekatan tersebut. Bahan x molar rendah x = 0.01, 0.02, 0.03, 0.04, 0.05, 0.06, 0.07, 0.08 dan 0.09 menunjukkan kesan kekonduksian dc yang ketara terutamanya pada suhu tinggi. Ini mungkin disebabkan oleh kesan peningkatan cas yang bergerak pada kawasan solidus cecair bahan. Bagaimanapun kesan kekonduksian dc pada sampel Pr(X)0.07 ialah disebabkan oleh penambahan kelincahan pelompatan kekonduksian ion bebas bagi mikrostrukturnya yang bersaiz butiran lebih kecil dengan kandungan Mn yang lebih tinggi.

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Finally, my foremost gratitude is for Agency Nuclear Malaysia for the scholarship study in Universiti Putra Malaysia.

I certify that a Thesis Examination Committee has met on 19 October 2009 to conduct the final examination of Nor Hayati binti Alias on her thesis entitled "Dielectric and Magnetic Characterization of $(La_{0.5-x}Pr_xBa_{0.5})(Mn_{0.5}Ti_{0.5})O_3$ Perovskite as a Multiferroic Material via Solid State Technique" 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 Master of Science.

Members of the Examination Committee are as follows:

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

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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.

NOR HAYATI BT. ALIAS

Date: 4 December 2009

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