UNIVERSITI PUTRA MALAYSIA

ELUCIDATION OF PARALLEL MICROSTRUCTURE AND EVOLUTION OF MAGNETIC PROPERTIES IN NICKEL ZINC FERRITE

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By

MUHAMMAD SYAZWAN BIN MUSTAFFA

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

November 2013

In appreciation of their love and sacrifices, this thesis is dedicated to my family especially my parents, MUSTAFFA BIN DOLLAH and NORMAH BT LUDIN and my brothers MUHD SYAZMER BIN MUSTAFFA and MUHD SYAZANI BIN MUSTAFFA and also my grandmother BUROK BT JOHAN who have been giving me full moral support throughout the years.

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

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Chairman: Associate Professor Mansor Hashim, PhD
Institute: Institute of Advanced Technology

The relationship between microstructural properties and magnetic characteristics of nickel zinc ferrite ($Ni_{0.6}Zn_{0.4}Fe_2O_4$) is the focus of interest in this research work. This interest has arisen from the fundamental scientific enquiry on the evolution of microstructure with the magnetic properties in between 1 nm-to-1 μm grain size region neglected by ferrites researchers for more than 80 years. Hence, here in this research work, we intend to track down the evolution of magnetic properties parallel to the microstructural changes in $Ni_{0.6}Zn_{0.4}Fe_2O_4$. Nickel zinc ferrite ($Ni_{0.6}Zn_{0.4}Fe_2O_4$) was prepared via High-energy ball milling (HEBM) using a SPEX8000D mill. Using single-sample sintering (SSS), the sample was repeatedly sintered with increasing temperature within the range 600^{o}C until 1200^{o}C with an increment of 25^{o}C in ambient air condition for 10 hours. After each sintering, the resulting changes in the materials were observed. The completion of solid state reaction was confirmed by using X-ray diffraction (XRD). Scanning Transmission

Electron Microscopy (STEM) was employed to confirm the particle size of the powder achieved. The evolution of microstructural properties of the sintered toroid was determined by using a Field Emission Scanning Electron Microscopy (FESEM) machine. The magnetic studies were carried out by using an Impedance Analyzer for AC response parameters, B–H Hysteresisgraph for B–H hysteresis loops and Precision Impedance Analyzer for Curie temperature measurements. The Archimedes principle was applied to determine the density of the toroid. The X-ray diffraction (XRD) patterns showed a single phase have been formed as early as 600°C and above and the intensity peaks increased with sintering temperature, indicating an increase in the degree of crystallinity. The morphological studies show a microstructural evolution (larger grain size) with the increased in sintering temperature. Grain size and density increased with increasing sintering temperature while the porosity decreased with increasing sintering temperature. An integrated analysis of phase, microstructural and hysteresis data points to the existence of three distinct shape-differentiated groups of B–H hysteresis loops which belong to the sample with weak, moderate and strong magnetism. The coercivity-vs-grain size plot reveals the critical single-domain-to-multidomain grain size to be about ~ 400 nm. The real permeability, μ' and loss factor, μ'', increase with grain size due to increase in sintering temperature. A scrutiny of the permeability components, μ' and μ'', also reveals three different groups which can be explained similarly by phase purity, microstructural data and crystallinity as in the B–H hysteresis case. The microstructural grain growth, as revealed for the first time by this research work is shown to be a process of multiple activation energy barriers. The Curie temperature gave the same value for each sintering temperature which means it remained

relatively stable and unaffected by the above evolutions. It is due to an intrinsic property which depends only on compositional stoichiometry and crystal structure.

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

PENJELASAN SELARI MIKROSTUKTUR DAN EVOLUSI SIFAT MAGNET DALAM FERIT NIKEL ZINK

Oleh

MUHAMMAD SYAZWAN BIN MUSTAFFA

November 2013

Pengerusi: Profesor Madya Mansor Hashim, PhD
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Hubungan antara sifat mikrostruktur dan ciri-ciri magnet ferit nikel zink ($Ni_{0.6}Zn_{0.4}Fe_2O_4$) adalah tumpuan penting dalam kerja-kerja penyelidikan ini. Kepentingan ini timbul daripada fakta bahawa siasatan saintifik asas evolusi mikrostruktur dengan sifat-sifat magnet bagi julat saiz butir 1 nm-ke-1 μm telah diabaikan oleh penyelidik ferit selama lebih daripada 80 tahun. Oleh itu, di sini, dalam kerja-kerja penyelidikan ini, kami berhasrat untuk menjejaki evolusi sifat magnet selari dengan perubahan mikrostruktur dalam $Ni_{0.6}Zn_{0.4}Fe_2O_4$. Ferit nikel zink ($Ni_{0.6}Zn_{0.4}Fe_2O_4$) telah disediakan melalui pengisar bebola berkuasa tinggi menggunakan pengisar SPEX8000D. Menggunakan pensinteran sampel tunggal (SSS), sampel telah berulang kali disinter dengan peningkatan suhu dalam julat 600^{o}C sehingga 1200^{o}C dengan kenaikan sebanyak 25^{o}C dalam persekitaran udara selama 10 jam. Selepas setiap pensinteran, hasil perubahan dalam bahan tersebut diperhatikan. Penyempurnaan tindakbalas keadaan pepejal diperiksa dengan

menggunakan pembelauan sinar-x (XRD). STEM telah digunakan untuk mengesahkan saiz zarah serbuk yang dicapai. Evolusi sifat-sifat mikrostruktur toroid yang disinter dikaji dengan menggunakan mesin FESEM. Kajian sifat-sifat magnet telah dijalankan dengan menggunakan Penganalisis Impedan/Bahan bagi parameter respons UA, B–H Histeresisgraf bagi gelung histerisis B–H dan Penganalisis tepat Impedan/Bahan bagi pengukuran suhu Curie. Prinsip Archimedes digunakan untuk menentukan ketumpatan toroid. Corak-corak pembelauan sinar-x menunjukkan fasa tunggal terbentuk seawal 600^{o}C ke atas dan puncak intensiti telah meningkat dengan suhu pensinteran yang menunjukkan peningkatan dalam darjah penghabluran. Kajian morfologi menunjukkan evolusi mikrostruktur (saiz butiran yang lebih besar) dengan peningkatan suhu pensinteran. Saiz butiran dan ketumpatan telah meningkat dengan peningkatan suhu pensinteran manakala keliangan menurun dengan peningkatan suhu pensinteran. Suatu analisis fasa, mikrostruktur, histerisis dan ketelapan bersepadu akan menunjukkan kepada kewujudan tiga kumpulan gelung histeresis B–H yang berasaskan bentuk, dengan pergantungan kepada suhu pensinteran, yang mewakili sampel yang mempunyai kemagnetan lemah, sederhana dan kuat. Graf medan-paksa-lawan-saiz butir memberikan nilai saiz butir kritikal bagi peralihan saiz butir domain-tunggal-ke-multidomain sebagai ~ 400 nm. Kebolehtelapan sebenar, μ' dan faktor kehilangan, μ'', meningkat dengan saiz butiran yang meningkat disebabkan oleh peningkatan dalam suhu pensinteran. Suatu penelitian parameter ketelapan, μ' dan μ'', menunjukkan parameter-parameter ini juga tergolong kepada tiga kumpulan yang boleh dijelaskan sama oleh ketepuan fasa, mikrostruktur dan penghabluran seperti dalam kes histerisis B–H. Pertumbuhan saiz butir dalam mikrostruktur, untuk pertama kalinya, telah didedahkan sebagai suatu proses yang melibatkan pelbagai nilai sawar tenaga keaktifan. Suhu Curie memberi nilai yang

sama bagi setiap suhu pensinteran yang bermakna ia kekal stabil dan tidak terjejas oleh evolusi di atas. Ia adalah disebabkan oleh sifat intrinsik yang bergantung hanya pada stoikiometri komposisi dan struktur kristal.

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APPROVAL

I certify that a Thesis Examination Committee has met on 25 November 2013 to conduct the final examination of Muhammad Syazwan bin Mustaffa on his thesis entitled "Elucidation of Parallel Microstructure and Evolution of Magnetic Properties in Nickel Zinc Ferrite" 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.

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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 Master of Science. The members of the Supervisory Committee were as follows:

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Date:

DECLARATION

Declaration by graduate student

I hereby confirm that:

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