



UNIVERSITI PUTRA MALAYSIA

***PARALLEL EVOLVING MORPHOLOGY, MAGNETIC PROPERTIES AND
THEIR RELATIONSHIPS IN $Ni_{0.5}Zn_{0.5}Fe_2O_4$***

ISMAYADI ISMAIL

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By

ISMAYADI ISMAIL

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in
Fulfillment of the Requirement for the Degree of Doctor of Philosophy**

January 2012

**In appreciation of their love and sacrifices, this thesis is dedicated to
Parents Haji ISMAIL AWANG and Hajah MEK ESAH AWANG, to my
beloved wife SAKINAH SHAMSUDIN and my three children, ZAFRAN
HAKIM, ARIF AMSYAR and HIJRIN BALQISH.**

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

PARALLEL EVOLVING MORPHOLOGY, MAGNETIC PROPERTIES AND THEIR RELATIONSHIPS IN $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$

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January 2012

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For more than seven past decades, the ferrite research literature has only very superficially dealt with the question of how the evolving microstructure of a ferrite material relates to its accompanying, resultant magnetic properties. The literature has only covered in great detail the answers for the case of ferrite materials obtained from final sintering. Thus, this work was a fresh attempt to critically track the evolution of magnetic properties parallel to the microstructural changes in bulk $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ samples and to relate the properties to the changes wherever possible. The study was divided into five parts. The first part involved two variables (milling time and ball to powder weight ratio (BPR)) of a mechanical alloying process where they were varied in order to study their effect on the magnetic properties of the material. The alloyed powders were used as starting powders for the rest of the research work. In the second

part, parallel sintering of a number of samples was carried out with sintering temperatures of 500 to 1400°C, subjecting one sample to only one particular sintering temperature. This was a multi-sample sintering process with 100°C increments. The third part dealt with higher-precision multi-sample sintering of several samples with sintering temperatures of 800 to 1000°C with much smaller increments of 25°C. The fourth part involved studies of the effect of soaking time on microstructural evolution and its influence on the magnetic properties. The last part carried out was similar to the second part (multi-sample), but it was run more carefully and critically with only one sample (single-sample) being subjected to various ascending sintering temperatures from 500 to 1400°C.

The results from first-part on the mechanical alloying parameters variation showed that there were no significant trends to relate the milling time and BPR with the permeability and losses of the material studied. After the samples were sintered at 1150°C, all the effects of the alloying process seemed to diminish. The results from the multi-sample sintering with the nanosized starting powders subjected to various sintering temperatures showed a clear development trend of the phase, morphology and magnetic properties of the samples. It is very interesting that the results revealed a critical region of sintering temperature for the development of magnetic properties which was observed at 800°C and 900°C with the sigmoid B-H curve shape taken to indicate a strong magnetic order. For the first time, this work has reported the evolution of the B-H hysteresis loops associated with the changes of magnetic states from paramagnetism to moderate ferromagnetism to strong ferromagnetism with microstructural changes. The results of the higher precision third part on the

relationship between ordered magnetism and the microstructure of the samples revealed a very startlingly systematic trend: a highly refined evolution trend covering a critical region of ordered magnetism which emerged and developed in step with morphological changes. Further work on the soaking time parameter was to study another possible way for the microstructure to influence the magnetic properties. The results showed a slow grain growth rate indicating a slow diffusion of atoms during the sintering process: it is believed that there was an increase in number of grain growth spots and these were the regions of mixed superparamagnetic and paramagnetic mass with ferromagnetic mass starting to dominate the samples. The last part of this work, carried out using single-sample sintering, also produced very gratifying results from the research point of view: the fascinating results from the single-sample sintering showed very systematically the evolution of microstructure-magnetic property relationships with a clarity superior to that shown by the multi-sample sintering.

Finally, after analysing the results and the observations of the work mentioned above, it is strongly believed that there are three factors found to sensitively influence the samples content of ordered magnetism –their ferrite-phase crystallinity degree, the number of grains above the critical grain size and large enough grains for domain wall accommodation. This research work has shed new light on the microstructure-magnetic properties evolution in ferrites.

Abstrak tesis yang dikemukakan kepada senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

**PERKEMBANGAN SEIRING EVOLUSI SIFAT-SIFAT MORFOLOGI,
MAGNETIK DAN HUBUNGANNYA DALAM $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$**

Oleh

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Bagi lebih daripada tujuh dekad yang lampau, literatur penyelidikan ferit hanya telah menangani secara amat cetek persoalan bagaimana sesuatu mikrostruktur ferit yang sedang mengalami evolusi berkaitan dengan sifat magnet yang terhasil seiring dengannya. Maka kerja ini adalah cubaan terbaru untuk mencerap secara kritikal evolusi ciri-ciri magnet seiring dengan perubahan mikrostruktur bagi sampel $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$. Kajian ini telah dibahagikan kepada lima bahagian. Bahagian pertama melibatkan dua pemboleh ubah (masa mengisar dan nisbah berat bola terhadap berat serbuk (BPR)) bagi proses pengalioian mekanik dimana pemboleh ubah tadi dipelbagaikan untuk mengkaji kesannya terhadap sifat magnet bahan. Dalam bahagian kedua, pensinteran selari bagi sampel-berbagai telah dilakukan dengan suhu pensinteran dari 500 sehingga 1400°C dengan setiap sampel hanya dikenakan satu suhu pensinteran. Ini adalah proses pensinteran pelbagai sampel dengan kenaikan 100°C. Bahagian ketiga berurusan dengan pensinteran selari pelbagai sampel yang lebih persis dengan suhu pensinteran dari 800

sehingga 1000°C dengan kenaikan yang jauh lebih kecil, 25°C . Bahagian keempat melibatkan kajian pada kesan masa rendaman pembakaran terhadap evolusi mikrostruktur dan pengaruhnya ke atas sifat magnet. Bahagian terakhir telah dibuat sama seperti bahagian kedua (pelbagai-sampel), tetapi dilakukan secara kritikal dengan hanya satu sampel (sampel-tunggal) dan dikenakan pelbagai suhu pensinteran secara menaik dari 500°C sehingga 1400°C .

Keputusan daripada bahagian pertama mengenai variasi parameter pengalioian mekanik menunjukkan bahawa tiada hala perubahan yang bermakna untuk mengaitkan masa mengisar dan BPR dengan keboleh-telapan dan 'kehilangan' bahan yang dikaji. Selepas sampel dibakar pada 1150°C , semua kesan-kesan proses pengalioian didapati hilang. Keputusan daripada pensinteran pelbagai-sampel dengan serbuk permulaan bersaiz nano dikenakan pelbagai suhu pensinteran menunjukkan perkembangan jelas fasa, morfologi dan sifat magnet bagi bahan. Adalah sangat menarik melihat keputusan-keputusan yang menyingkap suatu kawasan kritikal suhu pensinteran bagi pembinaan sifat magnet yang diperhatikan pada 800°C dan 900°C dengan bentuk sigmoid B-H diambil sebagai petunjuk tertib magnet yang kuat. Buat pertama kalinya kerja ini telah melaporkan evolusi gelung histeresis BH dikaitkan dengan perubahan keadaan dari keparamagnetan kepada keferomagnetan sederhana seterusnya kepada keferomagnetan kuat. Keputusan bahagian ketiga mengenai hubungan kemagnetan bertertib dengan mikrostruktur sampel menyingkap suatu hala perubahan sistematik yang amat menakjubkan: suatu hala evolusi yang amat halus yang meliputi suatu kawasan kritikal kemagnetan bertertib yang muncul dan berkembang seiring dengan perubahan-

perubahan morfologi. Kajian lanjut dibuat terhadap parameter masa rendaman pensinteran bagi mengkaji suatu lagi cara yang mungkin bagi mikrostruktur mempengaruhi sifat magnet. Keputusan-keputusan kajian ini menunjukkan kadar pembesaran butiran yang perlahan, menandakan resapan atom-atom semasa proses pensinteran: dipercayai terdapat peningkatan jumlah tompok-tompok pembesaran butiran dan ini adalah kawasan percampuran jisim superparamagnet dan paramagnet dengan jisim feromagnet yang mula mendominasi sampel. Bahagian terakhir kerja ini yang telah dijalankan dengan menggunakan pensinteran sampel-tunggal juga telah menghasilkan keputusan-keputusan yang amat memuaskan hati dari sudut penyelidikan: keputusan-keputusan menarik dari pembakaran sampel-tunggal menunjukkan secara amat sistematik evolusi hubungan mikrostruktur-sifat magnet dengan kejelasan yang lebih baik berbanding kes pensinteran pelbagai sampel.

Akhir sekali, selepas menganalisis keputusan dan pemerhatian kerja tadi, ianya amatlah diyakini bahawa terdapat tiga faktor yang secara sensitif mempengaruhi kandungan kemagnetan bertertib sampel iaitu darjah fasa kehabluran, jumlah butiran melepasi saiz kritikal butiran dan butiran yang cukup besar untuk penempatan dinding domain. Kerja penyelidikan ini telah menyumbangkan pengetahuan baharu tentang terhadap evolusi mikrostruktur-sifat magnet dalam bahan ferit.

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I cannot find the words for my parents **Haji Ismail Awang** and **Hajah Mek Esah Awang** “my Lord! Bestow on them Thy Mercy, even as they cherished me in childhood.”

I certify that a Thesis Examination Committee has met on 3rd January 2012 to conduct the final examination of Ismayadi Ismail on his thesis entitled “**Parallel Evolving Morphology, Magnetic Properties, and Their Relationships in $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$** ” in accordance with 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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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.

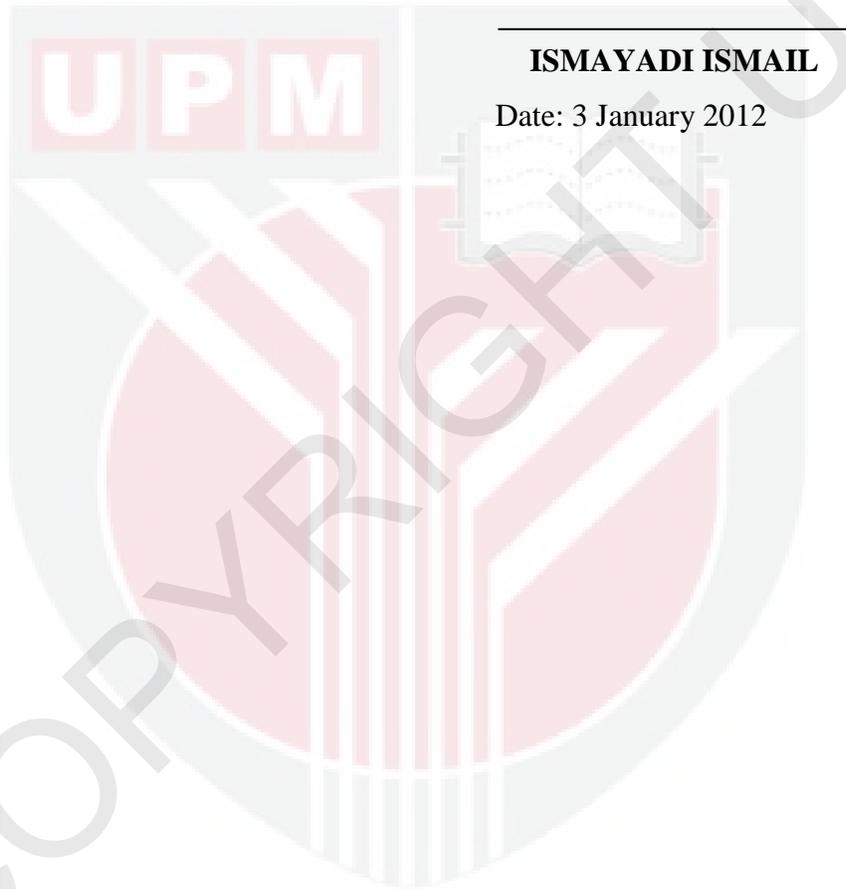


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LIST OF ABBREVIATIONS

XRD	x-ray diffraction
SEM	Scanning Electron Microscopy
EDX	Energy Dispersive X-ray
TEM	Transmission electron microscopy
wt %	Weight percent
hkl	Miller indices
MUT	Material under test
JCPDS	Joint Committee on Power Diffraction Standard
BPR	Ball-to-powder weight ratio
MA	Mechanical alloying
a.u	Arbitrary unit
Fig.	Figure
χ	Magnetic susceptibility
H	Magnetic field strength
H _C	Coercivity
M	Mass magnetization
M _R	Remanent magnetization
M _S	Saturation magnetization
σ_R	Specific remanent magnetization
σ_S	Specific saturation magnetization
θ_C	Curie temperature
θ_N	Néel temperature

2θ	2 theta degree
d_m	mean grain diameter
γ	magnetic domain wall energy proportional to the global anisotropy constant
M_{sv}	magnetization per unit volume
M_{sm}	magnetization per unit mass



CHAPTER 1

INTRODUCTION

1.1 Background of the Study

Ferrites are ceramic materials with magnetic properties which are mixed metal oxides containing iron oxide as their main component. They can be formed into permanent magnets for uses in motors, speakers and other electrical-mechanical energy conversion devices. They can also be formed into soft magnetic materials which are used as core materials for transformers, inductors and in microwave communication systems. Generally, ferrites are classified into three classes based on three different crystal types which are:

- 1) the spinel type, giving spinel ferrites,
- 2) the garnet type, giving garnet ferrites (as simply garnets)
- 3) the magnetoplumbite type, giving hexagonal ferrites.

Magnetically, the ferrites in categories 1) and 2) come under the class of “soft” ferrites while the ferrites in 3) belong to the class of “hard” ferrites. A soft magnetic material becomes magnetised by a relatively low applied magnetic field. When the applied field is removed, relatively low magnetism is retained in soft ferrites. Soft ferrites mostly

contain divalent or trivalent metal ions (nickel, zinc, manganese, yttrium, etc.), trivalent iron ions and divalent oxygen ions. Conversely, a high applied magnetic field is required for magnetizing hard ferrites. High remanent magnetism characterizes the properties of hard ferrites. They are prepared from iron oxide and barium oxide or strontium oxide.

Nickel Zinc ferrite is the most popular composition of soft ferrites. Due to the high resistivity and low eddy current losses and coercivity, ferrites of the nickel-zinc type are used in high frequency applications as a core material for power transformers and circuit inductors in the megahertz frequency region. They are more stable than the other types of ferrites, easily manufactured, low cost and have excellent and desirable magnetic properties. The properties of NiZn ferrite are sensitive to the compositional variability and the microstructure which is governed by the preparation process (Verma et al., 2005). In NiZn ferrites, the electrical and magnetic properties of ferrites depend on the stoichiometric composition. The nickel zinc ferrite with the well known composition of $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ is chosen in this study.

1.2 Microstructural-magnetic properties relationships of Ferrite Materials

Nanosized particles of the high-energy-milled spinel ferrites exhibit interesting physical and chemical properties markedly different from those of their bulk counterparts. The high-field magnetization irreversibility (Kodama et al., 1996), the variation of the Neel temperature with grain size (Chinnasamy et al., 2002), a high coercivity (Shi and Ding,

2001), and an altered (reduced (Lin et al., 1995) or enhanced (Oliver et al., 2000, Jiang et al., 1999 and Clark and Evans, 1997)) magnetic moments in comparison with the corresponding bulk materials have been observed in nanosize high-energy-milled ferrimagnetic spinels. Mechanical treatment of the spinel ferrites was found to be a useful activation method leading to an enhanced chemical reactivity of nanoscale powders (Sepelak et al., 1997). Nanostructured spinel ferrites prepared by high energy milling method are often inherently unstable owing to their small constituent sizes, nonequilibrium cation distribution, disordered spin configuration, and high chemical activity. Sintering of the milled spinel ferrites recrystallises the nanostructure and causes its transition from an excited metastable (activated) state into the low-energy crystalline state. During the process of sintering, the advantageous properties of the nanosize-milled spinel ferrites are mostly lost (Sepelak et al., 1998), thus an understanding of the relaxation mechanism of mechanically induced metastable states and of the thermal stability of nanostructure is necessary. A better understanding of the response of nanoscale spinel ferrites to changes in temperature is crucial not only for basic science (the development of an atomistic and microscopic theory of the mechanochemical processes) but also because of the technological high-temperature applications in catalysis, ferrofluids and information storage. To gain insight into the microstructural-magnetic property evolution relationships, this experimental work focuses on the study of the response of the fine nano-size starting powders of mechanically alloyed nickel zinc ferrite ($\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$) to changes in sintering temperature. Although extensive studies have recently been performed on the nanoscale-milled soft ferrites (Kodama et al., 1996, Chinnasamy et al., 2002, Sepelak et al., 2000, Oliver et al., 2000 and Jiang et

al., 1999), no measurements, of the thermally induced structural and magnetic evolutions in this metastable solid have been reported.

1.3 Problem Statement

A fundamental line of scientific enquiry has been neglected by ferrite and garnet researchers for more than 70 years: What would be the composition-microstructure relationships at various intermediate sintering conditions during the parallel evolutions of the morphology and the material properties? This research work intends to track carefully and critically the fundamental evolutions. Do the changes of microstructure affect the magnetic properties of the materials? How do magnetic properties evolve with the microstructure changes? What would happen to the ions of the materials parallel to the microstructure changes, do they also contribute to magnetization of the materials?

1.4 Objectives

The ultimate goal of this research is to critically track the evolution of magnetic properties parallel to the microstructural changes. Previous literatures mostly based on the micron-size starting powder and gradually were grown to the bulk solid samples. It is interesting to study the evolution of the magnetic properties with the nano-size starting powders and observe transition of paramagnetic to ferromagnetic behavior. This research work embarks on the following objectives:

- 1) To prepare $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ using mechanically alloyed nanoparticles.
- 2) To study the phase formation and crystallite size evolution of the as-prepared ferrite using XRD.
- 3) To study the effect of the sintering temperature on the microstructural evolution and magnetic properties of Ni-Zn ferrites.
- 4) To study the evolution of magnetic properties with microstructure changes.

1.5 Thesis Outline

General introduction of ferrite, microstructural-magnetic relationships and some research questions were discussed in chapter one while related literatures of the synthesis methods, composition tailoring, mechanical alloying and its important parameters, and some microstructural consideration on magnetic properties were reported in chapter two. Chapter three reported the basic theories as they affect ferrites and sintered materials. Specifically, the chapter reports the fundamentals of magnetization, the chemistry of spinel ferrites, sintering parameters and mechanical alloying process. The preoccupations in chapter four were methodologies employed for the preparations and the characteristics measurement of the as-prepared ferrite and sintered NiZn ferrite. The discussion of the obtained results of the as-prepared ferrites and the microstructure-magnetic properties forms chapter five. Chapter six summarized and concludes the research findings, in addition to some suggested recommendations. The list of his publications was attached at the end of the thesis, preceded by the author's biography appendices and references/bibliographies respectively.

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