UNIVERSITI PUTRA MALAYSIA

EFFECTS OF SINTERING TEMPERATURE ON MICROSTRUCTURAL AND DIELECTRIC PROPERTIES OF NICKEL-ZINC FERRITE PREPARED VIA SOL-GEL METHOD

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By

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Chairman: Assoc. Prof. Jumiah Hassan, PhD
Faculty: Science

In this work, $Ni_{0.5}Zn_{0.5}Fe_2O_4$ were synthesized via sol-gel method. The effect of sintering temperature on the microstructure and dielectric properties of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ were investigated. The ferrite samples were analyzed using X-Ray diffraction (XRD) to check the phases of the samples and to identify the formation of single phase $Ni_{0.5}Zn_{0.5}Fe_2O_4$. Field Emission Scanning Electron Microscope (FESEM) was used to get the magnified images of the morphology and microstructure of samples. The complex dielectrics of the samples were determined by HP 4192A LF Impedance Analyzer with setting of frequency range at 40 Hz up to 1 MHz while the measuring temperatures were fixed at 30°C and increased to 250°C. Keithley 236 Source which is connected to computer were set up for determining the DC conductivity of $Ni_{0.5}Zn_{0.5}Fe_2O_4$. Its measuring temperatures were fixed from 30°C to 250°C and the investigation involved the various sintering temperatures, starting at 600°C to 1100°C with increment of 50°C. The outcomes from XRD indicated that the single phase crystallization of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ started to form at initial sintering temperature of 600°C and they exhibited as a single phase cubic spinel structure with lattice parameter 8.60 Å. The higher of sintering temperature give an optimum crystallization, with greater density and lesser porosity of $Ni_{0.5}Zn_{0.5}Fe_2O_4$, hence contributing to produce more compacted sample with superior properties. The FESEM micrographs showed that the average grain size of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ nanoparticles was in the range of 59 nm to 199 nm with increasing sintering temperature from 600°C to 1100°C. This indicated that there was a grain growth occurred as the sintering temperature increased. Graphs of the frequency and sintering temperature dependence were tabulated for analyzing the variation of the dielectric constant and dielectric loss factor. The graphs trend show the dielectric constant and dielectric loss factor declined with greater frequency which is a standard dielectric properties of spinel ferrites. The Cole-Cole plots were tabulated to study the dielectric behavior of each sample. There are two primary relaxation

mechanisms generated while sintering every pellet sample, hence indicating a non-debye relaxation type. The DC conductivity of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ was analyzed to determine the conduction mechanism in the temperature reliance. As increasing sintering temperature, the values of electrical conductivity σ_{dc} increased representing the semiconducting behavior of the sample due to the upsurge the capability of the charge carriers to mobilize from grain-to-grain region in the samples.

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

KESAN SUHU PENSINTERAN PADA MIKROSTRUKTUR DAN SIFAT DIELEKTRIK NIKEL-ZINK FERRITE MELALUI KAEDAH SOL-GEL

Oleh

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Serbuk bersaiz nano-zarah $Ni_{0.5}Zn_{0.5}Fe_2O_4$ telah dihasilkan melalui kaedah sol-gel. Kesan pensinteran suhu ke atas mikrostruktur, sifat dielektrik dan asas elektrik pada $Ni_{0.5}Zn_{0.5}Fe_2O_4$ telah dikaji. Sampel ferit dianalisis menggunakan X-Ray pembelauan (XRD) dengan menyiasat ketulenan fasa sampel dan Field Pelepasan Mikroskop Imbasan Elektron (FESEM) bagi mendapatkan imej morfologi dan menganalisa bentuk mikrostruktur sampel. Sifat-sifat dielektrik sampel ditentukan menggunakan HP 4192A LF Impedance Analyzer dalam julat frekuensi 40 Hz hingga 1 MHz dan suhu pengukuran telah ditetapkan pada 30 °C sehingga 250 °C. DC kekonduksian pada nanozarah nikel zink ferit dikaji untuk menyiasat mekanisme pengaliran dan pergantungan suhu menggunakan komputerbersama mesin Keithley 236 Source. Suhu mengukur DC kekonduksian telah ditetapkan dari 30 ° C hingga 250 ° C dan penyiasatan itu melibatkan pelbagai suhu pensinteran, bermula pada 600 ° C hingga 1100 ° C dengan selang 50 ° C. Hasil XRD menunjukkan bahawa fasa penghabluran tunggal $Ni_{0.5}Zn_{0.5}Fe_2O_4$ mula terbentuk pada suhu pensinteran awal 600 ° C dan mereka dipamerkan sebagai satu fasa struktur spinel padu dengan kekisi parameter 8.60 Å. Suhu pensinteran yang tinggi memberikan penghabluran lebih halus pada sampel ferrite. Tambahan pula, peningkatan suhu pensinteran membuatkan sampel lebih tumpat dan kurang keliangan, dengan itu menyumbang kepada penghasilan sampel yang lebih padat dengan sifat-sifat unggul. Mikrograf FESEM menunjukkan kepelbagaian dalam taburan saiz zarah daripada 59 nm ke 199 nm dengan peningkatan suhu pensinteran dari 600 °C hingga 1100 °C. Ini menunjukkan mekanisme yang berbeza sepadan dengan pertumbuhan zarah berkenaan dengan suhu. Graf pergantungan frekuensi dengan suhu pensinteran telah dijadualkan untuk menganalisa perubahan yang berterusan dan faktor kapasiti dielektrik dan faktor kehilangan dielektrik. Corak graf menunjukkan kapasiti dielektrik dan faktor kehilangan dielektrik berkurangan dengan peningkatan frekuensi, selaras dengan sifat dielektrik taraf ferit spinel. The Cole-Cole plot telah dijadualkan untuk mengkaji tingkah laku dielektrik setiap sampel. Terdapat dua mekanisme kelonggaran utama dijanakan ketika pensinteran setiap sampel pelet, dengan itu

menunjukkan sampel tidak bersesuaian dengan Debye-jenis bersantai. DC kekonduksian nikel zink ferit dianalisis untuk menentukan mekanisme pengaliran arus dan pergantungan suhu. Nilai-nilai kekonduksian elektrik σ_{dc} meningkat dengan peningkatan suhu yang mewakili tingkah laku semikonduktor sampel kerana kerana peningkatan dalam pergerakan ion diaktifkan secara pengangkut pertuduhan terma.

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I certify that a Thesis Examination Committee has met on 13 June 2016 to conduct the final examination of Izzatil Ameerah binti Jaiah @ Jais on her thesis entitled "Effects of Sintering Temperature on Microstructural and Dielectric Properties of Nickel-Zinc Ferrite Prepared via Sol-Gel Method" 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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The 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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CHAPTER 1

INTRODUCTION

1.1 Background of Studies

In this day and age, high technology and communication becomes priority in daily individual life to pursue more efficient and productive profit. Improvements in researches are simultaneously pursued in semiconductors, dielectrics, ferroelectrics, ferrites and optical electronics. The sustained enhancements in these arenas correlated with improvements in ceramics, photonics, and optical waveguides (Hench and West, 1990). Variety of methodologies in nanoferrites' development have been studied and investigated from time to time, suitable for their broad range of applications, such as catalytic applications, drug delivery, sensors, magnetic resonance imaging, ferrofluids, microwave devices, antennas and RF frequency tune circuits. The innovations of ferrites' properties can be varied in method of preparations, sintering conditions and amount of elements. Nanoferrites exhibit distinctive properties as compared to the bulk counterparts.

Current study is attempting to apply top-down and bottom up approaches in order to synthesize nanostructure of nickel zinc ferrite, which is recognized as one of the members of soft magnetic ferrite. A top-down method defines a method which has to decompose a system for constructing the desired material. On the other hand, a bottom up method is a technique of piecing together, or combining the wreckages to custom a greater molecular system. The fundamental concept of synthesis method in nanotechnology is subjected to create molecular assemblages from the bottom up, by applying contemporary methods and tools to produce beneficial products. Via bottom up approach, the chemical reactions with adequate driving force can form atomic scale of particles into beyond micron level. Numerous of the latest processes are studied for assembling the atomic clusters to produce the preferred nano-sized material. In fact, the dielectric properties of ferrites are successively responsive to the synthesis condition since the each synthesize process involves the different cation distribution and microstructure variations. Hence, an appropriate process has to be selected to achieve desirable properties. Present research targets to improvise the simpler low temperature methods for providing ferrite nanoparticles in large, instead of producing by conventional techniques. Methods which consume lower temperature, lesser energy and more cost savings are the most desirable goals to get superior results with upgraded electrical and mechanical assets.

1.2 Ferrites

Ferrites are hard yet fragile structure in dark grey or black appearance. They are ceramic ferro magnetic materials which contain a mixed of iron (III) oxides with metal oxides, where iron (III) oxides are the main element with the general chemical formula is symbolized as $MeFe_2O_4$, where 'Me' is the divalent metal cation, Me^{+2}. The components manifest into the spinel structure where the cations engage with two types of sites, which are recognized as tetrahedral and octahedral sites. The capability to adjust the type of cations and their distribution in spinel ferrites can produce various exciting properties in applications. Ferrites can be crystallized into three groups which are hexagonal ferrites, garnet type ferrites and spinel ferrites. Hexagonal ferrites are used for permanent or hard ferrites applications, since they exhibit a high uni-axial magneto-crystalline anisotropy. Garnet type ferrites are distinctive magnetic ceramics which reveals the transparency requisite in magnetic optical uses. Spinel ferrites exhibit soft magnetic performance which are able to be magnetized or demagnetized smoothly by applying an external magnetic field. Spinel ferrites are the mainstream of recent attractively soft ferrites, which are characterized as a cubic crystal structure of the mineral spinel. Nickel zinc ferrites and manganese zinc ferrites are popular in the spinel ferrite family according to their high electrical resistivity and wide potential in altering the basic properties (Hench and West, 1990). Mn-Zn ferrites reveal greater initial magnetic permeability and saturation induction as compared to Ni-Zn ferrites. However, previous studies have shown that there are problems occurred by consuming Mn-Zn ferrites such as the untimely dielectric breakdown or mal-functioning because of their high electrical conductivity when operating below high frequencies and overheating by eddy currents as well.

In ceramic material, Ni-Zn ferrites are preferred for high frequency applications since they are more suitable for frequencies beyond 1 MHz and exhibit higher resistivity as compared to Mn-Zn ferrites ($MnZnFe_2O_4$). It is very convenient in high frequency applications as the suppression of eddy current is being a major attention (Kulikowski, 1984). The electrical properties, chemical composition and high sintering temperature process of this group of ferrites affect the grain size, grain boundaries and porosity of nickel zinc ferrites ($NiZnFe_2O_4$). $NiZnFe_2O_4$ are able to be handled in air, meanwhile $MnZnFe_2O_4$ required a redox conditions in the sintering chamber throughout the firing and cooling route. Nickel and zinc has very strong priority respectively at the tetrahedral and octahedral sites, which creates Ni-Fe a model inverse ferrite and Zn-Fe a normal ferrite. Those structure can occur as mixed spinel structures as well. Ni-Zn ferrites also reveal high electrical resistivity to avert eddy current and adequate magnetic permeability rather than the other ferrites. They are also more established than the other ferrites, simpler factory-made, low cost and have tremendous appropriate magnetic behaviors. Present attention is explored for nano sized nickel zinc ferrite in order to get more optimum results for different applications with the most effective manufacturing process.

1.3 Dielectric phenomena

Phenomena of dielectric was revealed in initial 600 BC, where the Greek philosopher Thales realized that there was a fascinating occurrence when a light piece of paper was attracted slightly into metal rod after chafing with a polyester sheet. It happened due to the electric polarization when an opposite charge on the paper surface adjacent the charge of rod tip, hence slightly producing an attraction force between them.

In the middle of 18^{th} century, the devotion in dielectric phenomena is not widespread recognized though the Leyden jar condenser was found to store charge in year of 1745

(Skilling, 1948). In 1837, Faraday introduced the existence of capacitance in the condenser. It was a correlation of the capacitance and condenser engaged to a dielectric material, subsequently free space and the specific inductive capacity is identified as permittivity.

The expansion of dielectrics studies specifically on the ferroelectric and the evolution of their special structures applications has directed to the manufacture of innovative categories of dielectric devices for radio-electronic and optical equipment. Dielectric properties are the most substantial behaviors of ferrites which obviously counted on the sintering time and temperature, type of material and technique of the process.

1.4 Problem statement

Instead of the excellent and desirable dielectric properties of nickel zinc ferrite, this ferrite was selected to be studied because of its behavior which are more stable than the other ferrites, easier to be sintered at the atmosphere control, plus easily manufactured and low cost. Composition of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ has been determined to be studied because of the dispassionate in investigating the compositional effect of nickel and iron in nickel zinc ferrite, dissimilar to the paper of Ajmal and Maqsood, 2007, which they has done research on $Ni_{1-x}Zn_xFe_2O_4$ ferrite system with (x=0.0, 0.2, 0.4, 0.6, 0.8, 1.0) by the standard solid state method. This research applied sol-gel method as the preparation technique of sample since this method has advantages of inexpensive precursors, a simple preparation method, resulting a homogeneous and nano-sized powder in bulk scale with high chemical homogeneity and low processing temperatures. Presently, many branches of communication and electronic manufacturing have been established in applications of ferrites. Common applications include audio and video recording tapes, motors, generators, recorders, computer disks, high frequency inductor and transformer cores (Moulson et al., 1990). Simultaneously, the fast developments of electronic industry demand higher density circuit in electronics components while primarily enhanced with multifunctional and high performance, smaller size and cost effectiveness of miniaturization of electronic chip components. Dielectric and electrical properties of materials gain extensive devotion recently and various operations have led dielectric materials to encounter the importance in multifunctional components (Mantese et al., 1996). It can reveal both high inductive and capacitive properties which have been determined to be an enabling solution to fabricate miniature filter and antenna, electro-magnetic interference (EMI) devices and others. It suggests investigating the microstructure-dielectric properties interactions at many intermediate sintering conditions in the parallel evolutions of the morphology and material properties. This research proposes to track the changes of dielectric properties parallel to the microstructural changes with multiple samples being sintered at different sintering temperatures and determine the best sintering temperature for this composition.

1.5 Research Objectives

The purpose of the present study is to investigate the changes of microstructural and dielectric properties of samples, where they are individually yielded at sintering temperature of 600 ˚C to 1100 °C, by increment of 50°C respectively. The objectives of this study are described as follows:

1. To prepare $Ni_{0.5}Zn_{0.5}Fe_2O_4$ nanoparticles using sol-gel method to study their microstructure-dielectric properties.

2. To determine the dielectric properties of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ at low frequency from 40 Hz to 1 MHz at measuring temperatures from room temperature to 250ºC.

3. To analyze the effect of the sintering temperature 600ºC to 1100°C on the microstructural evolution and dielectric properties of Ni-Zn ferrite.

REFERENCES

Adnan, A., Gul, S., and Ali, M. (2008). Content based image retrieval using geometrical shape of objects in image, *International Conference on Technology*. Peshawar, p. 222-225.

Alvani C., Ennas G., La Barbera A., Marongiu G., Padella F., Varsano F. (2005). Synthesis and characterization of nanocrystalline $MnFe_2O_4$ advances in thermochemical water splitting, *International Journal Hydrogen Energy*. 30(13-14), p. 1407-1411.

Ajmal, Muhammad and Asghari Maqsood. 2007. "Influence of Zinc Substitution on Structural and Electrical Properties of Ni1−xZnxFe2O4 Ferrites." *Materials Science and Engineering: B* 139(2-3):164–70.

Amador, D. R. and Torralba, J. M. (2003). Morphological and microstructural characterization of low alloying Fe powder obtained by mechanical attrition, *Journal Material Technology*, Vol. 143-144, p. 776-780.

Benjamin S. and Gilman P. S. (1983). *Metals Handbook*. (9^{th} ed.). ASM International, Ohio, Chapter 7, p. 722.

Borner I. and Eckert J. (1997). Nanostructure formation and steady state grain size of ball-milled iron powders. *Material Science Engineering*, 541, p. 226-228.

Broese Van Groenev A., Bongers P. F. and Stuyts A. L. (1968). Magnetism, microstructure and crystal chemistry of spinel ferrites, *Material Science Engineering*, 3, p. 317.

Chougule S. S. and Chougule B. K. (2008). Response of dielectric behavior on ferroelectric rich (x) $Ni_{0.8}Zn_{0.2}Fe_2O_4$+(1-x) PZT ME composites. *Material Chemistry Physics*, 108, p. 408.

Chen D. G., Tang X. G.,Tong J. J., Wu J. B., Jiang Y. P., Liu Q. X. (2011). Dielectric relaxation of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ Ceramics, *Journal Solid State Communications.*, 151, p. 1042-1044.

Coble R. L. (1961). Sintering crystalline solids. II. Experimental test of diffusion models in powder compact, *Journal Applied Physics*, 32, p. 793-799.

Cole K. S. and Cole R. H. (1941). Dispersion and Absorption in Dielectrics and Alternating Current Characteristics, *Journal Chemistry Physics*, 9, p. 341.
Costa, A.C.F.M., Tortella, E., Morelli, M.R., Kiminami, R.H.G.A. (2003). Synthesis microstructure and magnetic properties of Ni-Zn ferrites, *Journal Magnetic. Material*, 256, p. 174.

Cullity, B. D., (1978). *Elements of X-Ray Diffraction*, (2^{nd} ed.). Addison Wesley, UK.

Dias A. and Moreira R. L. (1999). Chemical, mechanical and dielectric properties after sintering of hydrothermal NiZn ferrites, *Material Letter,* 39, p. 69.

Dunitz J. D. and Orgel L. E. (1957). Electronic properties of transition metal oxides, Cation distribution amongst octahedral and tetrahedral sites, *Journal Physics Chemistry Solids,* 3, 318.

El-Sayed, A. M. (2002). Influence of zinc content on some properties of Ni-Zn ferrites, *Ceramic International.* 28, 363-367.

F. de Boer, Van Santen, J. H., and Verwey, E. J. W. (1937). Cation arrangement in spinels ferrites, *Procedure Royal Society,* London, 49, 59.

Fang H., Chen B., Jiang K., Sha J., Jiao Z., Zhang Q. and Zhang L. (1995). *Physics Status Solidi*, b 192, K11.

Fecht H. J., (1994). Proceedings of the NATO Advanced study institute on Nanophase materials-syntheses, properties and applications, *Kluwer academic publishers*, p. 125.

Forestier, H., Ann. Chemie Xe Series Tome. (1928). 9, 353.

Haijun Z. L., Zhichao L., Chenliang M., Xi Y., Liangying Z. and Mingzhong W. (2003). Preparation and microwave properties of Co-and Ti- doped barium ferrite by citrate sol-gel process, *Material Chemistry Physics,* 80, p. 129.

Hench, L. L. and West, J. K. (1990). Principles of electronic ceramics. *John Wiley & Sons.*

Heiks R. R. (1955). Relation of magnetic structure to electrical conductivity in NiO and related compounds, *Phyics Revision*, 99, 1232.

Heinicke G. (1984). Akademie Verlag, Berlin, Vol. 36, p. 400-401.

Homes C. C., Vogt T., Shapiro S. M., Wakimoto S., and Ramirez A. P. (2001). Optical response of high dielectric constant perovskite related oxide, *Science,* 293, p. 673.

Igarashi, H., Okazaki, K. (1977). Effects of porosity and grain size on the magnetic properties of NiZn ferrites, *Journal Ceramic Society,* 51(60).

Ismail, I.,Hashim M. (2014).Evolving microstructure, magnetic properties and phase transition in a mechanically alloyed $Ni_{0.5}Zn_{0.5}Fe_2O_4$ single sample, *Journal Magnetism and Magnetic Materials.,* 351, p. 16-24.

Iwauchi K. (1971). Interface Properties of Al_2O_3 Structure and Chararcteristics, *Journal Applied Physics*, 10, p. 1520.

Jonker G. H. (1959). Ferromagnetic compounds of manganese with perovskite structure, *Journal Physics Chemistry Solids,* 9, p. 165.

Jonscher A. K. (1983). Dielectric relaxation in solids, *Chelsea, Dielectric Press,* London. 3, 231.

Josyulu O. S. and Sobhanadri J. (1976). DC conductivity and dielectric behavior of cobalt zinc ferrites, *Journal Physics Statistic Solid,* 36K, p. 133.

Kulikowski, J. (1984). Soft magnetic ferrites development or stagnation, *Journal Magnetic Material*,41, 56

Koop C. G. (1951). On the dispersion of resistivity and dielectric constant of some semiconductors at audio frequencies, *Physics Revision*, 83, p. 121.

Kuhn E., (1984). "*Powder Metallurgy", ASM Handbook,* Vol. 7, ASM International: Material Park, OH. p. 56-70.

Louh, R., Reynolds III, T. G. and Buchanan, R. C. (2004). *Ferrite Ceramics in Ceramic materials for electrocnics* (3^{rd} ed.). Marcel Dekker inc., New York.

Maensiri, S., Masingboon, C., Boomchom, B., Seraphin, S. (2007). A simple route to synthesize nickel ferrite ($NiFe_2O_4$) nanoparticles using egg white, *Scripta Materialia,* vol. 56, p. 797.

Mantese, J. V., Micheli, A. L., Dungan, D.F., Geyer, R.G., BakerJarvis & Grosvenor, J. (1996). Applicability of effective medium theory to ferroelectric/ferrimagentic composites with composition and frequency-dependent complex permittivities and permeabilities. *Journal Applied Physics.* 79(3):1655-1660.

Mangalaraja R. V., Ramam K. V. S., Ravi J., Carlos P., Camurri. (2008). Synthesis of nanocrystalline yttria by microwave assisted citrate gel decomposition technique, *Journal Material Process Technology*, 197, p. 292-295.

Mathur R. B.,Chatterjee S., and Singh B. P. (2008). Growth of carbon nanotubes on carbon fibre substrates to produce hybrid/phenolic composites with improved mechanical properties. *Compos Science Technology*, 68, p. 1608-1615.

Maxwell J. C. (1982). A Treatise on Electricity and Magnetism, *Clarendon Press,* Oxford. Vol. 2, (3rd ed.).

McClure D. S., (1957). First Singlet-Singlet transition, *Journal Physics. Chemistry. Solids*, 3, 311.

Mohan R. M., Amara C. E., Cunningham D. A., Duffin J. (1999). Department of Physiology, Universiti of Toronto,Ontario, Canada, 115, 23-33.

Mohan G. R., Ravinder D., (1999). Dielectric properties of polycrystalline mixed nickel-zinc ferrites, *Material Letter,* 40, p. 39-45.

Moltgen S. (1952). *Z. Angew. Physics*, 4, 216.

Moulson, A. J. and Herbert, J.M. (1990). *Electroceramics: Material Process Applied,* (1st ed.). Chapman & Hall, London.

Mu C., Chen N., Pan X., Shen X. and Gu X. (2008). Preparation and microwave absorption properties of barium ferrite nanorods, *Material Letter* 62, p. 840.

Murr L. E. (1982). Electron and Ion Microscopy and Microanalysis, Principles and Applicatons, *Mc. Graw Hill Inc.* New York. 544.

Naik A. B. and Powar J. I. (1985). Electric and magnetic behavior of ferrites, *Indian Journal Pure and Applied Physics,* 23, 436.

Nazlan R., Ismail I., (2014). Complex permeability, Curie temperature and activation energy as a function of microstructure evolution in a mechanically alloyed $Y_3Fe_5O_{12}$ single sample, *Journal Basic and Applied Science* 8(3), p. 474-482.

Parvatheeswara Rao B., Subba Rao P. S. V. and Rao K. H. (1977). *IEEE Transnational. Magnetic,* 33, 4454.

Plocharski J. and Wieczorek W. (1988). PEO based composite solidelectrolyte containing Nasicon, *Solid State Ionics,* 28-30, p. 970-983.

Pradhan, A. K., Hammel, K. R., Deramus, R., Pollatsek, A., Noyce, D. A., Fisher, D. L. (2005). *Human Factors,* 47(4), 840-852.

Ponpandian N., Balaya P., and Narayanasamy A. (2002). *Journal Physics Condenser. Matter,* 14, 3221.

Ramalho, M. A. F., Gama, L., Antonio, S. G., Paiva-Santos, Miola, E. J., Kiminami, R. H. G. A., and Costa, A. C. F. M. (2007). X-Ray diffraction and Mossbauer spectra of nickel ferrite prepared by combustion reaction. *Journal of Material Science*, 42, 3603-3606.

Ranga Mohan G. and Ravinder D. (1999). *Material Letter,* 40, 39-45.

Rahman, M. N. (2003). Ceramic Processing and Sintering. *CRC Press.*

Ravindranathan, P., Patil, K. C. (1987). *Journal Material Science,* 3261(22).

Rao, B. P., S. V. S. Rao and K. H. Prao. (1997). Densification, grain growth and microstructure of Ni-Zn ferrites, *Journal Physics* Iv France.

Rao P. B.C., Setty S. P., Paldouglas S., Venkateswara rao B. (2011). *International Journal Engineering Technology Science,* Vol 2(3), 238-240.

Rezlescu N. and Rezlescu E. (1974). Dielectric properties of copper containing ferrites, *Journal Physics Status Solid,* 23, 575.

Rezlescu N., Sachelarie L., Rezlescu E., C-L., SavaandP. Popa, D. (2003). Influence of PbO on microstructure and properties of a NiZn ferrite. *Ceramics International*, 29, pp. 107-111.

Samokhvalov, A. A. and Rustamov, A. G. (1964). *Soviet Physics Solid State, 6, 749.*

Sivakumar, N., Narayanasamy, A., Jeyadevan, B., Justin Joseyphus, R., and Venkateswaran, C. (2008). *Journal Physics D: Applied Physics*, 41**,** 2451.

Skilling, H.H. (1948). *Fundamental of electric waves* (2nd ed.). J. Wiley &Son Incorporated.

Smith J. and Wijn H. P. J. (1959). Ferrite, Philips Technical Library, Eindhoven, The Netherlands, p, 137.

Snelling, E. C. (1969). *Soft Ferrites: Properties and Application* (1st ed.). Life Books Ltd., London.

Snoek, J. L. (1947). New developments in ferromagnetic materials. *Elsevier Publishing Co.* New York-Amsterdam.

Soohoo R. F. (1960). Theory and Application of Ferrites, *Prentice-Hall*, p. 30.

Tayal D. C. (1988). Electricity and Magnetism, *Himalaya Publishers.* 5, p. 372.

Thakur, A. and Singh, M. (2003). Preparation and characterization of nanosize $Mn_{0.4}Zn_{0.6}Fe_2O_4$ ferrite by citrate precursor method, *Ceramics International*, Vol. 29, p. 505.

Trivedi U. N.,Chantbar M. C., Modi K. B. and Joshi H. H. (2005). *India Journal Pure Applied Physics*, Vol. 43, p. 668-690.

Upadhyay Trupti, Upadhyay R. V. & Mehta R. V. (1997). Characterization of temperature sensitive magnetic fluid, *Physics Revision Book*, 55(9), 5585.

Verma, A., Goel, T. C., Mendiratta, R.G., Alam, M. I. (1999). Dielectric properties of NiZn ferrites prepared by the citrate precursor, *Material Science Engineering Book*, 60, p. 156.

Van Uitert, L. G. (1956). High resistivity nickel ferrites the effect of minor additions, *Process IRE*, 44, 1294.

Verwey E. J. W., De Boer F., and Van Santen J. H. (1948). Cation arrangement in spinels, *Journal Chemistry Physics.* 16, 1091.

Watt I. M. (1997). *The principles and practice of electron microscopy.* (2nd ed.). Cambridge University Press, p. 263-302.

Wagner K. W. (1913). On the theory of imperfect dielectrics, *Journal Physics* Vol. 345, p. 817-855.