UNIVERSITI PUTRA MALAYSIA

ELECTROMAGNETIC CHARACTERIZATION OF Sm-YIG AND Sm-YIG-PVDF COMPOSITES PREPARED USING MODIFIED CONVENTIONAL MIXING OXIDE TECHNIQUE

RAMADAN MASOUD AL-HABASHI

ITMA 2009 3

ELECTROMAGNETIC CHARACTERIZATION OF Sm-YIG AND Sm-YIG-PVDF COMPOSITES PREPARED USING MODIFIED CONVENTIONAL MIXING OXIDE TECHNIQUE

RAMADAN MASOUD AL-HABASHI

DOCTOR OF PHILOSOPHY
UNIVERSITI PUTRA MALAYSIA

2009

ELECTROMAGNETIC CHARACTERIZATION OF Sm-YIG AND Sm-YIG-PVDF COMPOSITES PREPARED USING MODIFIED CONVENTIONAL MIXING OXIDE TECHNIQUE

By

RAMADAN MASOUD AL-HABASHI

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

September 2009

Appendixes

DEDICATION

This thesis is dedicated to my son, daughters, wife and parents; to whom their true love and support motivated my success.

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the Degree of Doctor of Philosophy

ELECTROMAGNETIC CHARACTERIZATION OF Sm-YIG AND Sm-YIG-PVDF COMPOSITES PREPARED USING MODIFIED CONVENTIONAL MIXING OXIDE TECHNIQUE

By

RAMADAN MASOUD AL-HABASHI

September 2009

Chairman: Zulkifly Bin Abbas, PhD
Institute: Advanced Technology

Samarium substituted-yttrium iron garnet (Sm-YIG) nanoparticles were fabricated via a modified conventional mixing oxides (MCMO) method according to the $Y_{3-x}Sm_xFe_5O_{12}$ system ($0 \leq x \leq 3$). In this research, utilization of an organic compound (ethanol) and metal oxides in conjunction with mixing the reactants directly without adding water are the key techniques of this method. Using ethanol solution instead of water could produce nanoparticles with better homogeneity and smoother surface structure. Single-phase garnet structure of Sm-YIG nanoparticles was produced at 1350 ^{0}C sintering temperature with an average particle size ranged from 25 to 39 nm. XRD results of Sm-YIG samples at x = 2 and 2.5, presented some unknown peaks which speculated to, the time or/and sintering temperature is/are not enough to form the garnet structure phase of the samples. The true density values of 5.245 and 6.221 g.cm^{-3} were calculated for pure yttrium iron garnet (YIG, x=0) and samarium iron garnet (SmIG, x=3) samples, respectively which reached around 99% of the theoretical density of the samples.

Real permittivities of the Sm-YIG samples presented almost flat values ranged from 7 to 10 with loss factors around 0.1 to 0.3, for YIG (x=0) and SmIG (x=3) respectively, within 10 MHz to 1 GHz frequency range. The real permeability value 19.5 is presented by pure YIG at 13.4 MHz and declined rapidly to be around 2 at 1 GHz, and decreased with increasing x. The higher permeability resulted in lower permittivity and vice versa for all the Sm-YIG samples.

This work was also carried out to prepare the 10 wt% Sm-YIG in Poly-vinylidene-fluride (PVDF) composite samples and study their electromagnetic properties. Sm-YIG samples prepared via MCMO method, PVDF powder and Ethyl-methyl-ketone (MEK) were used to prepare such composites. High permittivities of composite samples observed at lower frequency range indicated to the heterogeneous conduction in the multiphase structure of the composites. The real permeabilities presented almost flat values through all the range of the frequency from 10 MHz to 1 GHz, with value of 1.06 at x=0 and 1.13 at x=3, for 10 wt% Sm-YIG loading in the composites. MCMO technique appears to be another alternative to the conventional (manufactured) technique, due to the decreasing of the particle size with better homogeneity, high purity, reduction of the cost, and high yield in a nano-scale product compared to other preparations techniques.

The numerical optimization method performed using MATLAB program is to estimate the effective complex permittivity and/or permeability of each component of the 10 wt% Sm-YIG-PVDF composite samples. It is found that, the optimum impedance values are very close to the measured ones for each composite. The optimized values of the complex permittivities and permeabilities for both

components [Sm-YIG and PVDF] are within the estimated ranges. The optimization process eliminated the difference between the measured impedance and the calculated one from Maxwell-Garnett (MG) formula via a specific objective function.

Results of a developed formula based on MG formula with a comparison of various theoretical models including the MG, Looyenga, Bruggeman and Sen-Scala-Cohen, have been carried out and discussed with comparisons to the measurements for the 10 wt% Sm-YIG-PVDF composite samples. This was to calculate the complex permittivity and permeability of such composite materials. The lowest mean error percentage values were detected from the developed MG formula for each composite, which was different from composite to composite depend on the mole fraction x. The developed MG model appears to add a new contribution to the theoretical models to calculate the effective permittivity and permeability of mixture ferrite-polymer materials, due to its accuracy as compared with others.

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

PENCIRIAN ELEKTROMAGNETIK KOMPOSIT Sm-YIG DAN Sm-YIG-PVDF MENGGUNAKAN TEKNIK DIMODIFIKASI KONVENSIONAL CAMPURAN OKSIDA

Oleh

RAMADAN MASOUD AL-HABASHI

September 2009

Pengerusi: Zulkifly Bin Abbas, PhD
Institut: Teknologi Maju

Zarah nano Samarium-YIG telah difabrikasikan melalui kaedah pengubahsuaian konvensional pencampuran oksida (MCMO) berdasarkan kepada sistem $Y_{3-x}Sm_xFe_5O_{12}$ $(0 \leq x \leq 3)$. Dalam kajian ini, penggunaan sebatian organik (etanol) dan oksida logam bersama dengan campuran reaktan secara terus tanpa penambahan air adalah kunci teknik untuk kaedah ini. Menggunakan larutan etanol menggantikan air boleh menghasilkan zarah nano dengan keseragaman yang lebih baik dan struktur permukaan yang lebih halus. Zarah nano Sm-YIG berstruktur garnet fasa tunggal telah dihasilkan pada suhu pemanasan 1350 ^{0}C dengan purata saiz zarah berjulat antara 25 ke 39 nm. Keputusan XRD bagi sampel Sm-YIG pada x=2 dan 2.5 menunjukkan beberapa puncak yang tak diketahui yang menggambarkan masa atau/dan suhu pemanasan adalah tidak mencukupi untuk membentuk struktur fasa garnet pada sampel. Nilai ketumpatan sebenar adalah 5.245 dan 6.221 g.cm^{-3} telah dikira untuk yttrium iron garnet (YIG, x=0) tulen dan samarium iron garnet (SmIG, x=3) masing-masing dimana mencapai 99% ketumpatan sampel secara teori.

Ketelusan sebenar bagi sampel Sm-YIG menunjukkan adalah kebanyakan nilai rata berjulat daripada 7 ke 10 dengan faktor kehilangan sekitar 0.1 ke 0.3 untuk YIG (x=0) dan SmIG (x=3) masing-masing dalam julat frekuensi 10 MHz ke 1 GHz. Nilai sebenar ketelapan 19.5 didapati dari YIG tulen pada 13.4 MHz dan berkurang dengan cepat sekitar 2 pada 1GHz dan berkurang dengan peningkatan jumlah x. Semakin tinggi nilai ketelapan menyebabkan semakin rendah nilai ketelusan dan sebaliknya untuk semua sampel Sm-YIG.

Kerja ini juga telah dilaksanakan untuk menyediakan 10 wt% Sm-YIG dalam sampel komposit Poly-vinylidene-fluride (PVDF) dan mengkaji sifat-sifat elektromagnetiknya. Sm-YIG sampel disediakan melalui kaedah MCMO, Serbuk PVDF dan Ethyl-methyl-ketone (MEK) telah digunakan untuk menyediakan komposit seperti ini. Ketelusan tinggi bagi sampel komposit telah diperhatikan pada julat frekuensi rendah menunjukkan kepada konduksi heterogen di dalam struktur multifasa komposit. Ketelapan sebenar menunjukkan kebanyakan nilai rata sepanjang kesemua julat untuk frequency dari 10 MHz hingga 1 GHz, dengan nilai 1.06 pada x=0 dan 1.13 pada x=3, bagi 10% Sm-YIG di dalam komposit. Ini menunjukkan percampuran baik campuran homogen komposit ferrite-polimer. Teknik MCMO juga adalah sebagai alternative lain bagi teknik konvensional (penghasilan), kerana dapatmenghasilkan pengurangan saiz zarah dengan homogen yang baik, ketulenan yang tinggi, pengurangan kos, dan hasil yang tinggi dalam produk skala-nano berbanding dengan teknik penghasilan yang lain.

Teknik pengoptimuman berangka telah dilakukan menggunakan program MATLAB. Ini adalah untuk menganggarkan ketelusan dan/atau ketelapan kompleks berkesan

bagi setiap komponen sampel komposit 10 wt% Sm-YIG di dalam PVDF. Didapati bahawa, nilai impedan optimum adalah sangat hampir dengan nilai yang diukur bagi setiap komposit. Nilai optimum bagi ketelusan dan ketelapan kompleks pada kedua-dua komponen [Sm-YIG dan PVDF] adalah didalam julat anggaran. Proses pengoptimuman telah menghapuskan perbezaan di antara impedan yang telah diukur dan yang dikira daripada formula Maxwell-Garnett (MG) dengan membezakan antara pelbagai model teori termasuk MG, Looyenga, Bruggeman dan Sen-Scala-Cohen, telah dijalankan dan dibincangkan dengan membezakan nilai pengukuran pada 10% Sm-YIG di dalam komposit sampel PVDF. Ini adalah untuk mengira ketelusan dan ketelapan kompleks bagi bahan komposit itu. Bagi setiap komposit, nilai purata ralat yang terendah telah didapati daripada formula MG yang telah dibangunkan, yang mana ini adalah berbeza daripada kebergantungan komposit kepada komposit pada pecahan mol x. Formula model MG yang telah dibangunakan dapat memberikan sumbangan baru kepada model teori untuk mengira ketelusan dan ketelapan efektif bagi bahan campuran polimer ferrite, berdasarkan kepada ketepatannya berbanding dengan yang lain.

ACKNOWLEDGEMENTS

First of all, great thanks to the Most Gracious and Most Merciful ALLAH (S.W.T) without his wish and help this work would not have been possible. I also would like to express the most sincere appreciation to those who made this work possible: Advisory members, Friends and Family.

I would like to thank Dr. Zulkify Abbas for providing me with the opportunity to complete my PhD. studies under his valuable guidance, for the many useful advice and discussions, for his constant encouragement and guidance, and for co-authoring and reviewing my publications, where his practical experience and technical knowledge made this research and those publications more interesting and relevant. Also special thanks extend to the supervisory committee members; Prof. Dr. Kaida Khalid and Prof. Dr. Mohd Maarof HJ Abd Moksin. I am grateful for their willingness to serve on my supervisory committee, constant encouragement, helpful advice and many fruitful discussions.

I also wish to extend my special appreciation to the staff of Institute Advanced Technology for their constant encouragement, kindness and helpful advice; the special thanks to the Program Manager, Electronic Material, AMNL, Assoc. Prof. Dr. Mansor Hashim for his kindness and helpful advice; and I would like to thank all my colleagues in AMNL for their helpful advice and many fruitful discussions. I also wish to extend my appreciation to Mrs. Rosnah Nawang and to the technician of AMNL, Mohd Kadri Masoud, for their kindness, support and for providing help whenever needed. Special thanks extend to Prof. Dr. Abd. Halim Shaari "Physics department" for his kindness, and providing help whenever needed.

I would like to express my appreciation and thanks to all who have support me during my life, in special, Mr. Alabyad, head of Plasma Physics Department, Tajoura Research Center, Libya. Ministry of Higher Education of Great Socialist People's Libyan Arab Jamahiriya is gratefully acknowledged.

I would like to extend my special thanks and appreciation to all my brothers, sisters and mother, brothers, sisters in laws for their unreserved encouragement throughout my study period and before.

Thanks and acknowledgements are meaningless if not extended to my parents who deserve my deepest appreciation. I am grateful for the countless sacrifices they made to ensure that I could pursue my dreams by always being there for me. Real and deepest thanks to them (May ALLAH bless and protect them and may live long and healthy). All praise and thanks words said to them will not be enough.

Lastly but not least, very very special thanks to my wife, son and daughters; confidante and true love. Their love, support, patience and encouragement are behind my success. They were always there to strengthen my soul during the darker hours and have always lightened-up my days.

I certify that a Thesis Examination Committee has met on 17 September 2009 to conduct the final examination of Ramadan Masoud Al-Habashi on his thesis entitled "Electromagnetic Characterization of Sm-YIG and Sm-YIG-PVDF Composites Prepared Using Modified Conventional Mixing Oxide 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 Doctor of Philosophy.

Members of the Examination Committee were as follows:

Wan Mohd Daud Wan Yusoff, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Azmi Zakaria, PhD
Professor
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Jumiah Hassan, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Ibrahim Abu Talib, PhD
Professor
Faculty of Science and Technology
Universiti Kebangsaan Malaysia
(External Examiner)

BUJANG KIM HUAT, PhD
Professor/ Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

Zulkifly Abbas, PhD
Lecturer
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Kaida Khalid, PhD
Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

Mohd Maarof Bin HJ Abd Moksin, PhD
Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

HASANAH MOHD GHAZALI, PhD
Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 16 November 2009

DECLARATION

I hereby 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.

_____

RAMADAN MASOUD AL-HABASHI

Date:

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES