



UNIVERSITI PUTRA MALAYSIA

***MAGNETIC, STRUCTURAL AND ELECTRICAL PROPERTIES OF
ALUMINIUM-SUBSTITUTED YTTRIUM IRON GARNET ($Y_3Fe_{5-x}Al_xO_{12}$
WITH $x=0, \dots, 3.0$) PREPARED VIA AUTO-COMBUSTION SOL-GEL
METHOD***

MUSA MAKIYU ABDULLAHI

FS 2016 66



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By

MUSA MAKIYYU ABDULLAHI

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

October 2016



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DEDICATION

To my beloved parents



Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the Degree of Master of Science

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By

MUSA MAKIYYU ABDULLAHI

October 2016

Chairman : Rabaaah Syahidah Azis, PhD
Faculty : Science

YIG ferrite material has been attracting attention due to its high electric resistivity, high radiation stability, comparatively low magnetization, narrowest ferromagnetic line width and consequent low loss. In this work, aluminum-substituted Yttrium iron garnet, ($Y_3Fe_{5-x}Al_xO_{12}$) powders, with $x = 0, 0.2, 0.4, 0.6$, to 3.0 were prepared using auto-combustion sol-gel method. The structural, magnetic and dielectric properties, at radio frequency range, were studied so as to contribute towards the study and improvement of microwave communication materials.

The X-ray diffraction (XRD) revealed the most representative peaks for YIG structure (400), (420) and (422) sharply reflected as matched by ICSD data. However, single garnet phase was found to be limited to $0.6 \leq x \leq 1.6$. The morphology of the samples, studied by Field Emission Scanning Electron Microscope (FESEM) shows decreasing grain sizes as Al concentration increases.

Magnetic properties of the samples were studied using vibrating sample magnetometer (VSM). Saturation magnetization, M_s was observed to decrease from 20.721 emu/g to 0.8112 emu/g as x increases from 0 to 3.0 , while remnance magnetization, M_r , decreased from 2.23 emu/g at $x = 0$, to 0 emu/g at $x = 1.8$ and above. The Coercivity, H_c changed non-linearly with x , having a maximum value of 39.00 Oe at $x = 0.8$ and a minimum value of 0 Oe at $x \geq 1.8$.

Dielectric properties of the samples were obtained at room temperature using Agilent impedance analyzer. At 1 MHz , the dielectric constant, ϵ_r of the samples decreases from its maximum value of 36.911 at $x = 0$ to its minimum value of 5.63 at $x = 0.6$. Dielectric loss tangent, $\tan \delta$ has a minimum value of 0.016 at $x = 3.0$ and a maximum value of 0.184 , while the pure sample has a value of 0.027 at 1 MHz . The conductivity decreased from $5.902 \times 10^{-5} \text{ Sm}^{-1}$ at $x = 0$ to its minimum value of 4.5×10^{-6} for $x = 0.4$, and increased thereafter, while the resistivity shows a reversed trend. The magnetic permeability, μ shows a decreasing trend increasing frequency. At 1 MHz ,

the value increased from 26.75 at $x = 0$ to 1263.12 at $x = 0.4$, while magnetic loss tangent, $\tan \mu$ decreased from 0.029 to 0.0096 as x increases from 0 to 0.4.

From the result, it shows that the Al-YIG materials have good magnetic and dielectric properties, $\mu_r > 1$, $\epsilon_r > 1$, $\tan \delta < 1$ and $\tan \mu < 1$. This is suitable for antenna miniaturization that can enhance the performance of radio wave communication devices.



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

**SIFAT MAGNET, STRUKTUR, DAN ELEKTRIK ALUMINIUM YTTRIUM
FERUM GARNET ($\text{Y}_3\text{Fe}_{5-x}\text{Al}_x\text{O}_{12}$, $x = 0, 0.2$, HINGGA 3.0) YANG
DISEDIAKAN MELALUI KAEDAH SOL-GEL AUTO-PEMBAKARAN**

Oleh

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Oktober 2016

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YIG ferit yang telah menarik perhatian disebabkan oleh kerintangan elektrik yang tinggi, kestabilan radiasi yang tinggi, pemagnetan yang rendah, jalur feromagnet yang sempit dan kehilangan yang rendah. Dalam kajian ini, aluminium-digantikan yttrium ferum garnet, ($\text{Y}_3\text{Fe}_{5-x}\text{Al}_x\text{O}_{12}$) dengan $x = 0, 0.2, 0.4, 0.6$, hingga 3.0 telah disediakan dengan menggunakan kaedah sol-gel auto-pembakaran. Sifat-sifat sample seperti struktur, mikrostruktur, magnet dan dielektrik, pada julat frekuensi radio, dikaji supaya dapat menyumbang ke arah kajian dan penambahbaikan bahan komunikasi gelombang mikro.

Pembelauan sinar-X (XRD) menunjukkan puncak yang ketara yang mewakili struktur YIG (400), (420) dan (422) dan dipadankan dengan data ICSD. Walau bagaimanapun, fasa garnet tunggal didapati terhadap kepada $0.6 \leq x \leq 1.6$. Morfologi sampel, telah dikaji oleh Field Pelepasan Mikroskop Imbasan Elektron (FESEM), menunjukkan penurunan saiz bijirin as peningkatan kepekatan.

Sifat-sifat magnet Al:YIG sampel telah diuji pada suhu bilik menggunakan sampel magnetometer bergetar (VSM). Pemagnetan tepu, M_s sampel diperhatikan menurun daripada 20.721 emu/g kepada 0.8112 emu/g apabila x bertambah 0-3.0, manakala pemagnetan baki M_r menurun daripada 2.23 emu/g di $x = 0$, ke 0 emu/g di $x = 1.8$ dan ke atas. Ini adalah disebabkan oleh kemagnetan yang lemah oleh paramagnet aluminium berbanding ferum feromagnetik. Coercivity, H_c berubah secara tidak linear dengan x , yang mempunyai nilai maksimum 39 Oe pada $x = 0.8$ dan nilai minimum sebanyak 0 Oe pada $x = 1.8$ dan ke atas.

Sifat dielektrik sampel diperolehi pada suhu bilik menggunakan Agilent Impedans penganalisis. Pada 1 MHz, pemalar dielektrik sampel berkurangan dari nilai maksimum daripada 36.911 pada $x = 0$ dengan nilai minimum sebanyak 5.63 pada $x = 0.6$. Kehilangan tangen dielektrik, $\tan \delta$ yang mempunyai nilai minimum 0.016 pada $x = 3.0$ dan nilai maksimum 0.184, manakala sampel tulen mempunyai nilai 0.027

pada 1 MHz. Kekonduksian menurun daripada $5.902 \times 10^{-5} \text{ Sm}^{-1}$ pada $x = 0$ untuk nilai minimum sebanyak 4.5×10^{-6} untuk $x = 0.4$, dan peningkatan selepas itu, manakala kerintangan menunjukkan trend terbalik. Kebolehtelapan magnet sampel menunjukkan aliran menurun dengan peningkatan dalam kekerapan. Pada 1 MHz, nilai meningkat daripada 26.75 pada $x = 0$ ke 1263.12 pada $x = 0.4$, manakala magnetic kehilangan tangen, $\mu \tan$ menurun 0.029-0.0096 sebagai x bertambah 0.0-0.4.

Dari keputusan itu, ia menunjukkan bahawa bahan-bahan Al-YIG mempunyai sifat-sifat magnet dan dielektrik yang baik, $\mu_r > 1$, $\epsilon_r > 1$, $\tan \delta < 1$ dan $\tan \mu < 1$. Ini sesuai untuk pengecilan antena yang boleh meningkatkan prestasi peranti komunikasi gelombang radio.



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This thesis was submitted to the Senate of the 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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LIST OF SYMBOLS AND ABBREVIATIONS

ρ	Resistivity
σ	Conductivity
μ	Permeability
ε	Permittivity
μ_r	Relative permeability
μ_0	Permeability free space
ε_r	Relative Permittivity
<i>AC</i>	Alternating current
<i>Al</i>	Aluminum
<i>DC</i>	Direct current
emu/g	Unit for magnetization emu/gram
<i>Fe</i>	Iron
FESEM	Field emission scanning electron microscope
FTIR	Fourier transform infrared
<i>G</i>	Conductance
H_c	Coercivity
HEBM	High Energy Ball milling
M_r	Remnant
M_s	Saturation magnetization
VSM	Vibrating sample magnetometer
XRD	X-ray diffraction
<i>Y</i>	Yttrium
<i>YIG</i>	Yttrium iron garnet



CHAPTER 1

INTRODUCTION

1.1 Ferrites

Ferrites are ferrimagnetic oxides of transition metals which have high electrical resistivity at room temperature (Vajargah *et al.*, 2007; Ghodake *et al.*, 2016). They are prepared for high frequency applications over their ferromagnetic counterparts, due to the minimum eddy current induction by an applied *ac* field. According to their crystal structure, ferrites are classified into spinels, garnet and hexagonal ferrites.

Spinel ferrites are ferromagnetic materials with general formula PQ_2X_4 , in which *P* represents divalent ions and *Q* represents trivalent ions. Originally, $MgAl_2O_4$ is the spinel material that occurs naturally with the same structure as ferromagnetic spinel materials (Morrish, 2001). Generally spinel ferrites can be formed by substituting the Aluminum ions with Fe^{3+} ions and the magnesium ions with any of the divalent ions (Fe^{2+} , Ni^{2+} , Co^{2+} , Mn^{2+} , Cu^{2+} etc). These forms uniquely high resistance ferrite materials that can be applied in microwave materials (Pardavi-Horvath, 2000). Spinel ferrites have a cubic, face-centered lattice structure surrounded by the predominant oxygen ions. The structure has two sub-lattices occupied by the cations. Tetrahedral (*A* site) is the smallest sub-lattice occupied by the magnetic cation and surrounded by four oxygen ions. The larger octahedral (*B* site) is formed by six oxygen ions surrounding the magnetic cation at the center. Eight tetrahedral *A* sites and sixteen octahedral *B* sites are occupied in a unit cell. In normal spinel structure, the eight ions occupying the tetrahedral site are divalent, *P*, while the sixteen ions occupying the octahedral sites are all trivalent, *Q*. However, if the tetrahedral site is occupied by eight trivalent ions and the octahedral by eight trivalent and eight divalent ions together, an inverse spinel structure is formed.

Natural garnets have an idealized general formula $A_3B_2Si_3O_{12}$, in which *A* and *B* can be Ca and Al ions respectively, with the possibility of several other compositions (Gerhard Winkler, 1981). Ferrimagnetic garnet belongs to a general formula $A_3B_5O_{12}$, where *A* is a rare-earth ion and *B* is a transition metal ion. It possesses body-centered cubic structure with three sub-lattices; dodecahedral *c* site, octahedral *a* site and tetrahedral *d* site. There are eight formula units in the crystal. In ferrimagnetic garnets all the cation sites are occupied by trivalent ions, which make them have a very low electron hopping and consequently, very high resistivity. The rare-earth metal cations occupy the dodecahedral site, while the tetrahedral and octahedral sites are occupied by three and two of the transition metal ions respectively. Due to their lower losses, garnets are preferred over spinel ferrites in most applications.

Hexagonal ferrites are oxide materials with a non-cubic crystal, hexagonal structure similar to that of the magnetoplumbite mineral material with chemical formula $PbFe_{7.5}Al_{0.5}Ti_{0.5}O_{19}$. The general formula for ferrimagnetic magnetoplumbite is

$\text{PO.6Fe}_2\text{O}_3$, where P can be Ba^{2+} , Sr^{2+} or Pb^{2+} . Hexaferrites are applied as permanent magnets due to their hard magnetic nature. They also have very small dielectric loss and are good insulating materials.

Ferrites are also classified into soft and hard ferrites, based on their magnetic behavior and applications.

Hard ferrites (also called ceramic materials) are those magnetic materials that cannot be easily magnetized or demagnetized due to their high coercivity (**Figure 1.1**). They are low cost magnetic oxides of iron, strontium or cobalt and are widely applied as permanent magnets in loud speakers, motors, refrigerator, etc.

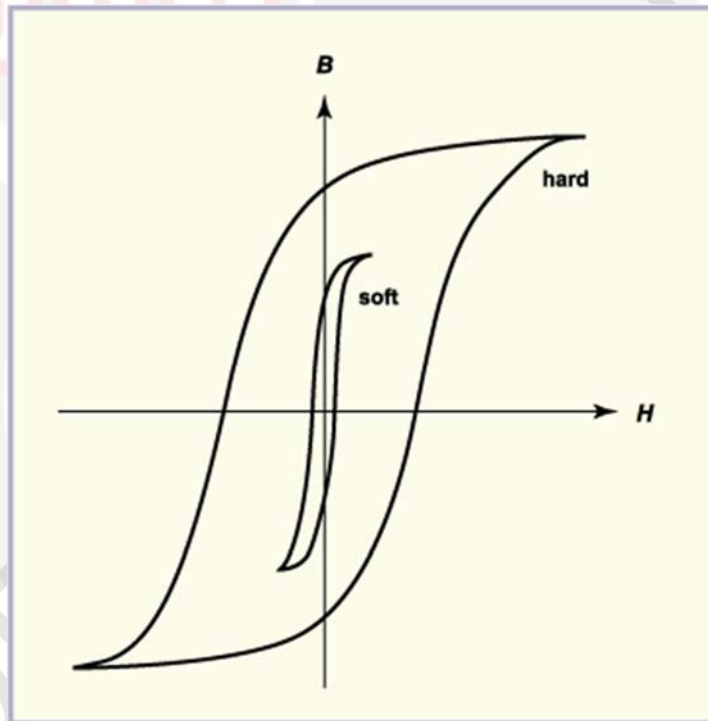


Figure 1.1 : Comparison between soft and hard magnetic materials

(Source: http://www.mtarr.co.uk/courses/topics/0238_ind/index.html)

Soft magnetic materials are materials with low coercivity and can be magnetized and demagnetized easily (**Figure 1.1**). Due to their low hysteresis loss and high resistivity, they are commonly used in transformers and electromagnetic cores. For high efficiency, especially in high-frequency application, some desirable properties of soft ferrites such as high saturation magnetization, low coercivity, high permeability and low hysteresis loss need to be improved. Soft ferrites are widely explored for high-frequency applications, in telephone signal transmitters, receivers and antennas.

1.2 Background of the study

The need for highly low loss materials is becoming a worldwide issue with the advances in satellite communication systems, base stations and antennas, most of which operate in microwave region. Microwave technology is advancing to higher frequency and bandwidth, which requires a miniaturized antenna as the size of the antenna is proportional to the wavelength. Thus, antenna downsizing is becoming an issue of interest in telecommunication devices, especially with the recent advances in terrestrial broadcasting in the ultra-high frequency (UHF) band (Fujii *et al.*, 2015).

Materials with high dielectric permittivity and thermal stability are required to achieve this miniaturization process as well as higher transmission and energetic efficiency (Paiva *et al.*, 2015). Ferrite materials have good magnetic and dielectric properties that make them potential antenna miniaturization materials (Sharma *et al.*, 2015).

The advancement of Yttrium iron garnet (YIG), $\text{Y}_3\text{Fe}_5\text{O}_{12}$, and gadolinium iron garnet (GdIG), $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ have significantly improved the performance of these microwave devices (Ramesh *et al.*, 2012). YIG is a ferrite material widely applied in microwave frequency devices. It has been attracting extensive research due to its wide variety of applications in optical devices and microwave communication components (Dolgova *et al.*, 2004; Akhtar *et al.*, 2014). This is because it is the magnetic material with the smallest magnetic resonance line width and, so, very low energy loss at microwave frequency (Rodziah *et al.*, 2012).

The efficiency of magnetic materials is enhanced by higher permeability. This can be achieved with an increased sintering temperature which produces larger grain size and, so, less grain boundaries. This will, in turn, make the system less porous with more domain wall movement which makes the permeability higher (Beitollahi and Hoor, 2003; Rodziah *et al.*, 2012).

1.3 Statement of the problem

High frequency electromagnetic materials have been receiving an increased attention, especially with the recent advancement in communication devices. Ferrites are the best materials for microwave applications due to their high electrical resistivity, ease of production and magnetic properties tailoring flexibilities (Pardavi-Horvath, 2000). Yttrium iron garnet (YIG) is a ferrimagnetic material widely applied in microwave electronic devices, phase shifters, wireless communications, magneto-optical devices, sensing and surveillance (Sadhana *et al.*, 2015). This is due to its promising magneto-dielectric properties (Paiva *et al.*, 2015). Sharma *et al.* (2016) reported that the ferrites' electromagnetic properties depend on the preparation technique, their constituent ions and metallic ions distribution on the tetrahedral and octahedral sites. Thus, ion substitution is believed to control their magnetic and magneto-dielectric properties. It was reported (Ata-Allah and Yehia, 2009) that some special electrical and magnetic functions are achieved by substitution of Iron (Fe) ions with some nonmagnetic trivalent ions such as Ga and Al ions in ferrites. In this work, Iron (Fe) ions were

substituted by Aluminum (Al) ions in YIG ferrite, forming Aluminum-substituted Yttrium iron garnet, Al-YIG, ($\text{Y}_3\text{Fe}_{5-x}\text{Al}_x\text{O}_{12}$) with $x = 0, 0.2, 0.4, 0.6$, to 3.0. Their structural, microstructural, magnetic and dielectric properties at radio frequency range were studied. This is because most of the reported works investigate YIG properties at microwave frequency range. The radio-wave frequency permeability was investigated using the permittivity-permeability relationship

$$Z = \sqrt{\frac{j\omega\mu}{\sigma + j\omega\varepsilon}} \quad \text{Eq. (1.1)}$$

1.4 Objectives of the study

Ferrites are magnetic oxide materials that possess good magneto electric properties suitable for high frequency applications. These properties, including high permeability high permittivity and low loss, are controlled by microstructural aspects (Sharma *et al.*, 2016). This research was conducted to study the structural, microstructural, magnetic and dielectric properties of Al-YIG ($\text{Y}_3\text{Fe}_{5-x}\text{Al}_x\text{O}_{12}$).

- a) To study the variation in morphology of aluminum-substituted yttrium iron garnet ($\text{Y}_3\text{Al}_x\text{Fe}_{5-x}\text{O}_{12}$) with $x = 0, 0.2, 0.4$, to 3.0, prepared via auto combustion sol-gel method.
- b) To study the effect of Al^{3+} substitution on structural, phase composition, electrical, magnetic and dielectric properties of the YIG samples

1.5 Significance of the study

The role of mobile and wireless communication in our daily lives cannot be over emphasized. This prompted the need to study and improve the performance of telecommunication devices that are widely applied in satellite communication systems, base station and antennas. Recent miniaturization of mobile communication devices accelerated the need for antenna downsizing (Mattei *et al.*, 2015). Ferrites are considered potential antenna miniaturization materials due to their high permeability, $\mu > 1$, permittivity $\varepsilon > 1$ and low loss, $\tan \delta < 0.01$ (Sharma *et al.*, 2015). YIG is a ferrite magnetic material with the smallest magnetic resonance line width and, so, very low energy loss at microwave frequency (Rodziah *et al.*, 2012). Rare earth iron-substituted garnets have been receiving scientific interests due to their applications in various fields. Aluminum-substituted garnets produces an interestingly new phenomena that attracts scientific interest recently (Kim *et al.*, 2003).

1.6 Outline of the thesis

This thesis is designed into six chapters. **Chapter 1** gives a general introduction of the research work, comprising of nanoparticles, ferrites, statement of the problem, objectives and significance of the study. **Chapter 2** reviews relevant literature on methods of preparing YIG. Structural, magnetic and electromagnetic/magneto-dielectric properties of YIG and its derived compounds were also reviewed. **Chapter**

3 covers theoretical aspects of the subject. It explains magnetism and its type, intrinsic and extrinsic magnetic properties and garnet structure. **Chapter 4** deals with materials and sample preparation method. The different characterizations measurements involved in the research are covered. **Chapter 5** presents the samples characterization results and discussion. Conclusions drawn from the research findings are given in **Chapter 6**.



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