DEVELOPMENT OF LOW LOSS HEXAFERRITE MATERIALS
FOR MICROWAVE APPLICATIONS

A Dissertation Presented
By
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to
The Department of Electrical and Computer Engineering
in partial fulfillment of the requirements
for the degree of

Doctor of Philosophy
in
Electrical Engineering
in the field of
Electromagnetics, Plasma, and Optics

Northeastern University
Boston, Massachusetts
May 2016
Hexaferrites have been widely used in microwave and millimeter wave devices as permanent magnets and as gyromagnetic materials, e.g., in circulators, filters, isolators, inductors, and phase shifters. As a critical component in radar and modern wireless communication systems, it is the microwave circulator that has drawn much attention. Many efforts have been made to design light and miniature circulators with self-biased ferrite materials. We report the magnetic and structural properties of a series of W-type barium hexaferrites of composition BaZn$_{2-x}$Co$_x$Fe$_{16}$O$_{27}$ where $x=0.15$, 0.20, and 0.25. The anisotropy field of these BaW ferrites decreased with the substitution of divalent Co ions, while, they maintained crystallographic c-axis texture. The measured anisotropy field was ~10 kOe, and a hysteresis loop squareness $M_d/M_s=79\%$ was obtained due to well-controlled grain size within the range of single domain scale. U-type barium hexaferrite thin films were deposited on (0001) sapphire substrates by pulsed laser deposition. The results indicate a measured anisotropy field of ~8 kOe, and the saturation magnetization ($4\pi M_s$) of 3.6 kG. More interestingly, an optimal post-deposition annealing of the films results in a strong (0, 0, n) crystallographic texture and a high squareness ($M_d/M_s=92\%$) out of the film plane. Furthermore, the highly self-biased ferrite films exhibited low FMR linewidth of ~200 Oe.

Improved performance and miniaturization are needed to meet the ever-increasing demands of devices used in ultra-high frequency (UHF), L-band, and S-band, which are of
particular interest in a variety of commercial and defense related applications. Utilizing materials possessing high permeability and permittivity with low magnetic losses is a promising solution. As a critical component in radar and modern wireless communication systems, antenna elements with compact size are constantly sought. Ferrite composites of the nominal composition $\text{Ba}_3\text{Co}_{2+x}\text{Ir}_x\text{Fe}_{24-2x}\text{O}_{41}$ were studied in order to achieve low magnetic and dielectric losses and equivalent permittivity and permeability over a frequency range of 0.3-1 GHz. Crystallographic structure was characterized by X-ray diffraction, which revealed a Z-type phase accompanied by increasing amounts of Y-type phase as the iridium amount was increased. The measured microwave dielectric and magnetic properties showed that the loss $\tan\delta_e$ and loss $\tan\delta_\mu$ were decreased by 80% and 90% at 0.8 GHz with the addition of iridium having $x = 0.12$ and 0.15, respectively. An effective medium approximation was adopted to analyze the composite ferrites having mixed phase structures. Moreover, adding $\text{Bi}_2\text{O}_3$ resulted in equivalent values of real permittivity and real permeability over the studied frequency range. The resultant data gives rise to low loss factors (i.e., $\tan\delta_e/\varepsilon' = 0.008$ and $\tan\delta_\mu/\mu' = 0.037$ at 0.8 GHz) while characteristic impedance was the same as that of free space impedance.
ACKNOWLEDGMENTS

I would like to thank my advisor, Professor Vincent G. Harris, for his guidance, sincere help and constant support during my study at Northeastern University. Professor Vincent G. Harris is a brilliant scientist, a selfless mentor, and a humorous gentleman. Especially, I really appreciate that my advisor offered me to join in his group when I feel hopeless at the first year of my graduate study, and also give all kinds of extreme support during the last period of my Ph.D. study when I am busy and anxious about my future. It’s my luck to have Professor Harris to be my advisor.

I would like to thank my co-advisor, Professor Yajie Chen, who is intelligent and knowledgeable. Professor Chen not only guides me with research ideas, but also helps me to solve every problem encountered during my graduate study. He teach me not only fundamental theories but also experimental method. He also helps me to revise academic articles and give suggestions about career pursuing. His sincere help would make a profound effect on my future work.

I would like to thank my committee member Professor Yongmin Liu, a nice professor, for his review and comments.

I would like to thank my group members for their friendly help with work and life: Dr. Xing Xing, Dr. Bolin Hu, Ms Hong Chang, Dr. Yunume Obi, Dr. Jianwei Wang, Dr. Steven Bennett, Dr. Scott Gillette, Dr. Trifon Fitchorov, Mr Alexander Sokolov, Ms Parisa Andalib, Ms Parisa Taheri, Mr Michael Geiler, and so on.
I would like to thank these visiting scholars in our group: Professor Xian Wang, Dr. Zongliang Zheng, and Professor Junliang Liu, who really help me a lot with my research.

Finally I would like to thank my family and my friends a lot for their everlasting care and support.

Last finally, to my dear husband, thank you for always being there.
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CHAPTER 1

INTRODUCTION

1.1 Magnetic hexagonal ferrite materials

1.1.1 Definition and classification of ferrites

Magnetic materials are ubiquitous throughout industry. Two well-known major branches of magnetic materials are metallic magnetic materials and ceramic mag-oxides. The ceramic oxide magnetic materials are called ferrites. There are three types of ferrite materials: spinels, garnets and hexaferrites [1-5]. The spinels and garnets exist as cubic crystal structures, whereas the hexaferrites exist as hexagonal crystal structures. Basically, different crystal structures will give them different magnetic, dielectric and mechanical properties. However they still possess some common properties: moderate to high values of magnetization, high permeability, high permittivity, high electrical resistivity, low dielectric and magnetic losses, high Curie temperature, good mechanical rigidity and strength, and stable chemical properties.

1.1.2 Spinels and garnets

Both spinels and garnets exist in cubic structures. Spinels have general formulation \( \text{A}^{2+}\text{B}^{3+}\text{O}^{2-}_4 \), where \( \text{A} \) indicates a divalent cation such as nickel (\( \text{Ni}^{2+} \)), cobalt (\( \text{Co}^{2+} \)), zinc (\( \text{Zn}^{2+} \)), copper (\( \text{Cu}^{2+} \)), and etc., \( \text{B} \) is usually a trivalent cation such as iron (\( \text{Fe}^{3+} \) is the most
The space group of spinels is $Fd\overline{3}m$ with 32 oxygen anions close packed into a face centered cubic (FCC) spinel lattice, among which two kinds of cations reside. Cations have either four-fold or six-fold coordination forming tetrahedra sublattices (A) and octahedra sublattices (B). For A sites, only 8 out of 64 available tetrahedral sites are occupied by cations, whereas for B sites, 16 out of 32 available octahedral sites are occupied [6]. The 3D crystal structure of spinels with tetrahedral and octahedral sublattices is shown in Fig. 1-1. The yellow tetrahedral elements and red octahedral elements are clearly seen in Fig. 1-1, all the oxygen anions appear as blue balls.

**Figure 1-1:** Schematic of spinel crystal structure with tetrahedral sites (A) and octahedral sites (B).
The intrinsic mechanism of magnetism of spinel crystal structure can be assumed as super-exchange interaction [7-10]. In the ground state super-exchange is a negative exchange interaction that results in the anti-alignment of cation spins (i.e., ferrimagnetic), which make the magnetization of ferrites much less than that of most 3d metallic alloys (i.e. Fe, Co, Ni), and the magnetism of metallic magnetic materials arises from direct exchange with the spins aligned parallel to each other (i.e., ferromagnetic) [6].

![Schematic of garnet crystal structure](image)

**Figure 1-2:** Schematic of garnet crystal structure with tetrahedral, octahedral, and dodecahedral sites.

Garnets have general formulation $\text{R}^{3+}_3\text{B}^{3+}_5\text{O}^{2-}_{12}$, where R indicates a rare earth trivalent cation such as Yttrium ($\text{Y}^{3+}$), Neodymium ($\text{Nd}^{3+}$), Gadolinium ($\text{Gd}^{3+}$), Samarium
(Sm$^{3+}$), and etc, B is usually a trivalent cation such as iron (Fe$^{3+}$ is the most popular one among others), aluminium (Al$^{3+}$), Chromium (Cr$^{3+}$), and etc. [11-12]. Yttrium iron garnet (YIG) is used in acoustic, microwave, optical, and magneto-optical applications, which make it very popular for industrial applications. For example as microwave filters, utility in solid-state lasers, Faraday rotators, and in nonlinear optical applications [6]. The space group of YIG is $Ia_3d$, where trivalent Fe ions occupy two octahedral and three tetrahedral sites, and the yttrium ions occupy dodecahedral sites (a 12-sided distorted polyhedral) that are irregularly surrounded by 8 oxygen ions in a distorted cube [6]. The 3D crystal structure of garnets with tetrahedral, octahedral, and dodecahedral sublattices is shown in Fig. 1-2. The red tetrahedral and octahedral lattices, and the purple dodecahedral lattices are clearly seen in Fig. 1-2, all the oxygen anions are given as blue balls.

The magnetism of garnets also comes from the super-exchange as in the spinel systems. The Fe$^{3+}$ ions on octahedral and tetrahedral sites have antiparallel spins, therefore are coupled ferrimagnetically. The excess Fe$^{3+}$ ions from tetrahedral sites contribute to the net magnetic moment. YIG shows relatively low saturation magnetization and the magnetic anisotropy field, which also means a low ferromagnetic resonance frequency. Thus when YIG is used for microwave applications, a biased magnetic field is usually introduced. Since all the available octahedral and tetrahedral sites in garnet lattice are occupied by metallic cations, this completely filled structure makes garnets easy to have incredible magnetic and chemical properties, one of them is extremely small FMR linewidth [13], i.e., very low magnetic loss, which makes garnet widely used in modern communication and microwave applications.
1.1.3 **Hexaferrites**

Hexagonal ferrites were first discovered in the 1950s. Hexaferrites have a hexagonal crystal structure. The existing six types of hexaferrite structures are designated as: M, Y, Z, W, X and U. The general formulation of hexaferrites is $M_x M_{12}(Fe_{2}O_{3})_y$, the ratios of $x/y$ for M, Y, Z, W, X and U are 1:6, 1:3, 1:4, 1:8, 1:7 and 2:9, respectively [5-6, 14-16]. M in the chemical formula of hexaferrites represents the ions of Ba, Sr, Pb, Ca, La, etc., and Me is usually a transition element (Zn, Mg, Mn, Co, etc.). As is the same in the spinel structures, the substitution of Fe$^{3+}$ ions can be other trivalent cations such as Al$^{3+}$, Cr$^{3+}$, etc.

**Table 1-1**: Summarized details of six types of hexaferrites with molecular formula, lattice parameters and stacking sequences (use barium ferrites here as example).

<table>
<thead>
<tr>
<th>Hexaferrite type</th>
<th>Formula</th>
<th>$\sim a$ (Å)</th>
<th>$\sim c$ (Å)</th>
<th>Stacking sequences</th>
</tr>
</thead>
<tbody>
<tr>
<td>M</td>
<td>$BaFe_{12}O_{19}$</td>
<td>5.88</td>
<td>23</td>
<td>RSR<em>S</em></td>
</tr>
<tr>
<td>Y</td>
<td>$Ba_2Me_2Fe_{12}O_{22}$</td>
<td>5.88</td>
<td>43</td>
<td>TST’S’T’”'S”''</td>
</tr>
<tr>
<td>Z</td>
<td>$Ba_3Me_2Fe_{24}O_{41}$</td>
<td>5.88</td>
<td>52</td>
<td>RSTSR<em>S</em>T<em>S</em></td>
</tr>
<tr>
<td>W</td>
<td>$BaMe_2Fe_{16}O_{27}$</td>
<td>5.88</td>
<td>33</td>
<td>RSSR<em>S</em>S*</td>
</tr>
<tr>
<td>X</td>
<td>$BaMeFe_{28}O_{46}$</td>
<td>5.88</td>
<td>84</td>
<td>RSR<em>S</em>S*</td>
</tr>
<tr>
<td>U</td>
<td>$Ba_3Me_2Fe_{36}O_{60}$</td>
<td>5.88</td>
<td>113</td>
<td>RSR<em>S</em>TS*</td>
</tr>
</tbody>
</table>
For the hexagonal structure, two lattice parameters are used to describe it: a (the width of the hexagonal plane) and c (the height of the unit cell), here all six type hexaferrites have a similar \( a \approx 5.88 \, \text{Å} \), however lattice parameter c varies among the six types, i.e., the heights of the six type hexaferrite crystal unit are different from each other. All six types of hexagonal crystal structure can be described as an alternating stacks of three fundamental blocks: S, R and T. They can be also built with S*, R* and T*, which come from the 180° rotation of S, R and T blocks around c-axis of the crystal lattices. All the above information is summarized in Table. 1-1.

Figure 1-3: Schematic of M type hexaferrite crystal structure with polyhedral perspective.
M type hexaferrites have the simplest stacking structures among all six hexaferrites. Barium (BaFe$_{12}$O$_{19}$) or Strontium (SrFe$_{12}$O$_{19}$) substituted hexaferrites are the most popular and well-studied M type hexaferrites. Barium M type hexaferrites have the magnetoplumbite structure, with a space group of $Pb3/mmc$ and a stacking of RSR*S*.

The unit consists of 10 oxygen layers with 38 oxygen ions, 24 iron ions and 2 Ba ions in total. Two Ba cations locate in the R and R* blocks, which is separated by an S block with two oxygen layers, and between every two Ba ions there are four oxygen layers. The iron ions are arranged on five sites: three octahedral sites (2$a$, 12$k$ and 4/2), one tetrahedral site (4$f1$) and one trigonal bipyramidal (TBP) site (2$b$). The Schematic of M type hexaferrite crystal structure in polyhedral perspective is shown in Fig. 1-3.

Y type hexaferrite crystal structures belong to $R3m$ space group, and they have a stacking sequence of TST’$S$’T”$S$”, one T block and one S block, followed with a T’ and S’ block (T’ and S’ represent the 120° rotation around c-axis of the corresponding T and S blocks), and another following T’’ and S’’ block (T’’ and S’’ represent the 240° rotation around c-axis of the corresponding T and S blocks).

The molecular formula of barium Y type ferrites is Ba$_2$Me$_2$Fe$_{12}$O$_{22}$, where Me is usually divalent transition metal ions with the occupation of both tetrahedral and octahedral sites. The Y type hexaferrites have a c-plane magnetic anisotropy, which indicates a preferred magnetization perpendicular to the c-axis when applied with external magnetic field. The Schematic of Y type hexaferrite crystal structure in polyhedral perspective is shown in Fig. 1-4.
**Figure 1-4:** Schematic of Y type hexaferrite crystal structure with polyhedral perspective.
Figure 1-5: Schematic of Z type hexaferrite crystal structure with polyhedral perspective.
Z type hexaferrite crystal structures have a space group with \( \text{Pb3/mmc} \), and a stacking sequence of RSTSR*S*T*S*, which can also be seen as the combination of M and Y type hexaferrites. The molecular formula of barium M type ferrites is \( \text{Ba}_2\text{Me}_2\text{Fe}_{24}\text{O}_{41} \), where Me is usually divalent transition metal ions with the occupation of tetrahedral, octahedral and trigonal bipyramidal sites. The Schematic of Y type hexaferrite crystal structure in polyhedral perspective is shown in Fig. 1-5. Since Y type hexaferrites have a c-plane magnetic anisotropy while M type hexaferrites have a c-axis anisotropy, the preferred magnetization of Z type hexaferrites various with different metal substitution. It is found that most divalent metal substituted Z type hexaferrites have magnetic anisotropy perpendicular to the c-axis, however \( \text{Co}_2\text{Z} \) hexaferrites have a c-axis anisotropy. Both \( \text{Co}_2\text{Y} \) and \( \text{Co}_2\text{Z} \) hexaferrites have extremely small coercivity, this soft magnetic property plus planar magnetization preference make it possible to be used for magnetic core of inductors, antenna substrates and other microwave components like spinel ferrites. Besides, compared to spinel ferrites \( \text{Co}_2\text{Y} \) and \( \text{Co}_2\text{Z} \) hexaferrites have large magnetic anisotropy fields, which will provide the opportunity of microwave devices applications at GHz frequency range.

W type hexaferrite crystal structures belong to \( \text{Pb3/mmc} \) space group, and they have a stacking sequence of RSSR*S*S*, which is very similar to the stacking sequences of M type hexaferrites. The molecular formula of barium W type ferrites is \( \text{BaMe}_2\text{Fe}_{16}\text{O}_{27} \), where Me is usually divalent transition metal ions with the occupation of tetrahedral, octahedral and trigonal bipyramidal sites. The Schematic of W type hexaferrite crystal structure in polyhedral perspective is shown in Fig. 1-6. Like Z type hexaferrites, the preferred magnetization of W type hexaferrites various with different metal substitution.
Most of the divalent metal substituted W type hexaferrites (i.e. Zn$_2$W, Ni$_2$W and Fe$_2$W) hexaferrites have magnetic anisotropy perpendicular to the c-axis, however Co$_2$W hexaferrites have a c-axis anisotropy. The Schematic of X type and U type hexaferrite crystal structures in polyhedral perspective are shown in Fig. 1-7 and Fig. 1-8, respectively.

**Figure 1-6:** Schematic of W type hexaferrite crystal structure with polyhedral perspective.
Figure 1-7: Schematic of X type hexaferrite crystal structure with polyhedral perspective.
Figure 1-8: Schematic of U type hexaferrite crystal structure with polyhedral perspective.
1.2 Microwave ferrite devices

Ferrite materials have two major advantages compared to metallic magnetic materials: one is that ferrite material have low eddy current losses due to the nature that they are a group of insulating magnetic oxides [6]; the other is the nonreciprocal property, which can regulate the microwave signal direction of the transmission and reception by its intrinsic magnetization preference along crystal planes or axes. Besides, ferrite materials possess moderate to high values of magnetization, high permeability, moderate to high permittivity, high electrical resistivity, and low losses at high frequency range [6], which make them good candidate for modern communication and microwave devices applications with high quality of electromagnetic signal process and low power consumption.

As stated, spinel ferrites have high saturation magnetization, low magnetic anisotropy field, high initial permeability and high resistivity. Ni-Zn spinel ferrites have been considered as good soft magnetic materials for microwave application below 1GHz, and they are widely used in power electronics and RF applications as inductors and transforms due to the merits of relatively high permeability and electrical resistivity [17]. Since the ferromagnetic resonance (FMR) frequency is determined in part by magnetic anisotropy field, spinel ferrites have low ferromagnetic resonance frequency due to reason of low magnetic anisotropy field. Thus the operating frequency related with ferromagnetic resonance frequency of spinel ferrites based microwave devices are also low, general speaking below 1GHz, moreover a biased external magnetic field may be applied to shift the ferromagnetic resonance frequency for some microwave devices applications to higher frequency [14].
Garnet ferrites have low ferromagnetic resonance frequency (FMR) linewidth, relative low saturation magnetization and low magnetic anisotropy field compared to other ferrite materials. Yttrium iron garnets (YIG) have extremely low FMR linewidth among all the garnet ferrites, namely very low magnetic loss, which makes them one of the most industrially important garnets for microwave devices applications. However, like spinel ferrites, garnet ferrites possess low magnetic anisotropy field compared to hexaferrites bring the limitation to themselves for high frequency range (above 1 GHz) microwave applications, or an external permanent magnet is needed for biased field.

Hexaferrites have moderate to high saturation magnetization, high magnetic anisotropy field, high electrical resistivity, moderate Curie temperature, and moderate magnetic linewidth. Due to the unique property of high magnetic anisotropy field compared with spinel and garnet ferrites, hexaferrites make themselves good candidate to microwave and millimeter-wave applications at high frequency range above 1 GHz. Barium M type hexaferrites (BaFe\textsubscript{12}O\textsubscript{19}, BaM) is well known engineering ferrite materials as commercial used permanent magnets, magnetic recording materials, and microwave devices. BaM hexaferrites have hard magnetic properties, corresponding to a large remnant magnetization. The high uniaxial magnetocrystalline anisotropy field of BaM hexaferrites acts as a large effective internal magnetic field of the microwave circulator, which provides the possibility of eliminating the external permanent magnets and helps to achieve the miniaturization of size, weight lightening, cost saving [18]. Co substituted Y and Z type hexaferrites (Ba\textsubscript{2}Co\textsubscript{2}Fe\textsubscript{12}O\textsubscript{22}, Co\textsubscript{2}Y and Ba\textsubscript{3}Co\textsubscript{2}Fe\textsubscript{24}O\textsubscript{41}, Co\textsubscript{2}Z, respectively) are another two promising commercial microwave products at high frequency range, due to the properties of in-plane magnetic anisotropy, high permeability, and high resistivity. The soft
magnetic properties and high ferromagnetic resonance frequency are unbeatable features for microwave applications as antenna substrates, inductor cores and electromagnetic band gap (EBG) structures at high or even millimeter-wave frequency range [19].

1.2.1 **Microwave circulators and isolators**

![Diagram](Image)

**Figure 1-9:** Schematic of a stripline Y-junction circulator (a), electric field distribution in the stripline Y-junction circulator (b), and Simplified block diagram of a T/R module (c) [6].

The circulator is a passive non-reciprocal element used to regulate the direction of the transmission signal in microwave and wireless communication systems, with which only one antenna is need instead of two for both transmitting and receiving at duplex
working mode. The schematic of a Y-junction circulator in stripline configuration is shown in Fig. 1-9 (a), two ferrite disks with one layer stripline circuit form the sandwich structure, and two permanent magnets are used as external biased field. The electric field distribution in the stripline Y-junction circuits simulated by finite element methods is given in Fig. 1-9 (b). Port 2 is the output with respect to port 1 as input, whereas port 3 is isolated simultaneously. The Y-junction circulator is often used in transmit-receive (T/R) modules of phased array radar systems [20], a simplified block diagram of the T/R modules is shown in Fig. 1-9 (c).

Theoretical calculation of stripline Y-junction circulators was solved approximately by Bosma in 1964 [21]. The first-order circulation resonance condition is given by Eqn. 1-1, as \( \omega \sqrt{\varepsilon_0 \mu_0} = 2\pi / \lambda \), \( \lambda \) is the wavelength of free space, Eqn. 1-1 can be replaced by Eqn. 1-2, which is straightforward and more practical for engineering circulator device design. Here R is the radius of the ferrite disk, and \( \lambda \) corresponds to the operating wavelength. Other details about the determination of insertion angles for the coupling stripline, frequency dependence, and bandwidth enhancement were also studied by Bosma [21].

\[
x = kR = 1.84 \quad (1 - 1)
\]

\[
R = \frac{1.84}{2\pi \sqrt{\mu_{eff} \varepsilon_r}} \quad (1 - 2)
\]

With the development of millimeter-wave monolithic integrated circuit technology towards the trend of the size reduction, the design of the microwave circulator also face an intensive challenge, it is known that microstrip planar junction circulator configuration offers the lowest overall size and cost [22]. Recent results of novel planar circulator at high frequency range will be examined next.
A design of planar circulator based on magnetic nanowired substrate (MNWS) operating at millimeter wave frequency range is proposed by A. Saib et al [23]. The schematic of the designed MNWS circulator is shown in Fig. 1-10. The magnetic substrate used here consists of an array of parallel ferromagnetic nanowires electrodeposited into a porous polycarbonate, this kind of design has two advantages: one is that the circulator is planar and easy to integrate, the second is that this design is self-biased, i.e. no need of external biased magnetic field. An insertion loss less than 2 dB with higher than 45 dB isolation is achieved with MNWS circulator at 26 GHz.

**Figure 1-10:** Schematic of the designed MNWS circulator: polycarbonate membrane bulk under the circulator disk with nanowired composite attached as sidewall.
1.2.2 Magneto-dielectric antenna substrate

The magneto-dielectric materials can miniaturize both the EBG structures and antenna by the same factor however using moderate values of permittivity and permeability ($n = \sqrt{\varepsilon'\mu'}$) compared to the dielectric-only high permittivity material [19].

The use of $\varepsilon'$ identical to $\mu'$ has two benefits: for multilayer EBG structures, it produces better band-gap rejection levels [19]; for antennas, it allows for ease of impedance matching over a much wider bandwidth. Furthermore, an electrically small antenna may be matched to any frequency by a proper selection of geometry, the use of matching circuits, etc., but it may sacrifice antenna efficiency. Thus, the magneto-dielectric materials are of significant interest in antenna design and materials research. More importantly, the use of $\varepsilon'$ identical to $\mu'$ gives more flexibility of antenna design in order to simply fabrication and efficiency of antennas.

The designs of artificial magneto-dielectric materials with a prescribed permittivity and permeability values obtained by proper arrangement of available non-magnetic materials are very popular recently [24-26].
Figure 1-11: (a) Geometry of the magneto-dielectric embedded-circuit medium. (b) Effective constitutive parameters of the meta-material.

A design of magneto-dielectric meta-substrate constructed of periodic resonant loop circuits embedded in a low dielectric host medium with the capability of providing both permittivity and permeability material at any frequency of interest is proposed by H. Mosallaei et al [24]. The schematic of the periodic structure and the effective parameters are shown in Fig. 1-11.

Another design of arrays of split-ring resonators (SRRs) used as an artificial magneto-dielectric substrate to reduce the resonant frequency of patch antenna is proposed by M. Karkkainen et al [25]. In this work a wider impedance band for an antenna with stacked SRRs embedded in a low-permittivity substrate is achieved, as compared with one in a higher-permittivity substrate without SRRs. The schematic of the patch antenna with stacked SRRs structure is shown in Fig. 1-12.
However all the designs of artificial magneto-dielectric materials have complicated three dimension structures, which is not a good choice for commercial production. The exploration of natural magneto-dielectric materials without artificial structures is attracting in the view of engineering applications.

Figure 1-12: The patch antenna under investigation (a) as seen from above and (b) with $x$-oriented SRR stacks, side view.
1.3 Low loss microwave ferrites

Low loss ferrite materials are widely used in microwave elements such as circulators, isolators, antenna substrates, inductors and so on. Magnetic, dielectric, and conductor losses are all contribute in microwave devices, which are represented by insertion loss as a critical parameters of device performance [4].

The dielectric loss due to damping of the vibrating electric dipole moments and conductor loss are expressed by the imaginary part of the permittivity. Magnetic loss comes from the magnetic damping related with ferromagnetic resonance and domain wall motion. The eddy current, induced by changed magnetic flux, also give rise to nonnegligible loss in oxide magnetic medium due to the larger skin depth compared to metals. Since the magnitude of eddy current is inversely proportional to the resistivity of the material, high resistivity ferrites are wanted to achieve low loss. Some extrinsic situations will also affect the ferromagnetic resonance linewidth, such as crystal defects from the growth, random local anisotropy, porosity, grain boundaries, surface morphology, and so on [2,27].

As mentioned before, Hexaferrites make themselves good candidate to microwave and millimeter-wave applications at high frequency range above 1 GHz due to the unique property of high magnetic anisotropy field, one of which is self-biased microwave circulator. Traditional circulators need external biased magnetic field for operation, which are bulky and costly to fabricate [28]. Self-biased circulator are light and compatible. Low loss self-biased circulators are attracting for microwave engineering.

In the case that magnetic ferrites materials are used in microwave devices due to their nonreciprocal character, the width of the ferromagnetic resonance curve play an
important role in indicating the loss. Self-biased ferrite materials are usually polycrystalline, which has larger ferromagnetic linewidth than those of single crystal.

Figure 1-13: SEM images of low loss hexaferrite thick film (a) Surface and (b) cross section [28].
A barium hexaferrite thick film with narrow ferromagnetic resonance linewidth was given by Yajie Chen et al [28]. The measured ferromagnetic linewidth was 310 Oe, compared with that in LPE films of 27 Oe at 56 GHz [29]. It is given that the density of the thick film is 88%, i.e., porosity of 12%, which is also confirmed by SEM images in Fig. 1-13. Since about 70% of the total linewidth is attributed to porosity in the thick film, Polycrystalline materials being inevitable with porous, give rise to a significantly broader ferromagnetic linewidth compared to the effects of anisotropy, even for 99% dense ferrites [30]. However increasing the film density and enhancing the alignment of grains with the c-axes perpendicular to the film plane will help to further optimize the ferromagnetic linewidth.

The low loss self-biased polycrystalline M-type Sc-doped ferrites with lower anisotropy fields for X-band applications are proposed by Yajie Chen et al [31]. The density of the samples are measured to be around 92%, which is confirmed by the SEM images in Fig. 1-14. The in-plane ferromagnetic linewidth is measured to be 550 and 830 Oe, respectively, corresponding to two resonance frequencies at different applied fields. It is estimated that the porosity contributes 440 to 900 Oe to the width of linewidth, compared to the 32 Oe from the random orientation of grains and 50 Oe from the intrinsic linewidth, respectively. It is seen that the polycrystalline system with inevitable porosity has a rather broader linewidth, which can be optimize by thermal treatment or other growth control method [31].
Figure 1-14: SEM images of low loss Sc-doped hexaferrite sample (a) Surface and (b) cross section [31].

Ferrite materials with high permeability and low magnetic loss are ideal candidates for microwave high frequency antenna substrates and inductors applications. Hexaferrites, such as Co$_2$Z and Co$_2$W, with planar anisotropy, have relative high permeability and higher resonance frequency compared to spinel structures.
Figure 1-15: Permeability spectra of W type hexaferrites show domain wall resonance and natural resonance [32].

In polycrystalline ferrites, the frequency dispersion of permeability is affected by two mainly resonance mechanisms, namely, the natural resonance and domain wall resonance [32]. Since microwave devices with ferrite materials like antennas and inductors are operated in off-resonance condition, the operating frequency and magnetic loss of the
devices are affected by the resonance frequency and resonance linewidth of the ferrite materials, respectively. Permeability spectra of W type hexaferrites show domain wall resonance and natural resonance are given in Fig. 1-15 [32]. The peak at lower frequency represents the domain wall resonance, while the higher one is the natural resonance. It’s seen that the domain wall resonance make the peak of imaginary permeability spectra wider than just with natural resonance, which lower the cut off frequency for applications and also worsen the loss at frequency range near resonance point. Decreasing the grain sizes of ferrite materials will reduce the domain wall resonance effect, which will help to lower the magnetic loss. However it may at the same decrease the real permeability.

1.4 Reference


CHAPTER 2

SELF-BIASED W-TYPE HEXAFERRITES FOR X-BAND MICROWAVE APPLICATIONS

2.1 Introduction to X-band microwave ferrites

Barium hexaferrites have been widely used in microwave and millimeter wave devices as permanent magnets and as gyromagnetic materials, e.g., in circulators, filters, isolators, inductors, and phase shifters [1-2]. As a critical component in radar and modern wireless communication systems, it is the microwave circulator that has drawn much attention. Many efforts have been made to design light and miniature circulators with self-biased ferrite materials [3]. We have successfully demonstrated self-biased circulators that operate at Ku-band based on M-type hexaferrites [4]. However, demonstrating self-biased circulators at lower operating frequencies, i.e. X-band, is more difficult due to the need for lower magnetic anisotropy fields and correspondingly lower FMR frequencies. In our previous work, we have developed a processing scheme in which self-biased M-type barium ferrite films with thicknesses up to 500 microns have been produced that operate at U band [5]. Furthermore, we made progresses in the synthesis of M-type barium ferrite with Sc doping, which is responsible for reducing the anisotropy field and FMR frequency [6].
Both W-type and M-type barium ferrite have high saturation magnetization, high Curie temperature, and c-axis easy magnetic anisotropy. However, Ni or Zn substituted W-type barium ferrites have magnetic anisotropy fields of ~12 kOe and FMR frequencies that are much lower than those of the unsubstituted M-type ferrites. Furthermore, substituting Co ions can alter the alignment of the anisotropy field from c axis to c plane [7-10]. Additionally, the Ni-Co or Zn-Co substituted W-type barium ferrites have anisotropy fields from 4 kOe to 12 kOe with c-axis texture, respectively. In recent years, most studies of W-type barium ferrites have focused on exploring its soft magnetic properties for low reflection materials at microwave frequency [11-16].

### 2.2 Zn substituted W-type barium ferrites

In this work, a series of BaZn\(_{2-x}\)Co\(_x\)Fe\(_{16}\)O\(_{27}\) W-type barium ferrites (BaW) were prepared and studied for their structure and magnetic properties. In particular, we successfully demonstrated that BaW hexaferrites might be made to possess self-biased properties over relatively low microwave frequencies in the X-band.

#### 2.2.1 Preparation of crystallographically textured W-type ferrites

Polycrystalline BaW ferrites having composition BaZn\(_{2-x}\)Co\(_x\)Fe\(_{16}\)O\(_{27}\) where x=0.15, 0.20, and 0.25 were prepared by a conventional ceramic process that entailed repeated steps of firing and ball milling, as shown in Fig. 2-1. Starting materials of BaCO\(_3\), ZnO, Co\(_2\)O\(_4\), and Fe\(_2\)O\(_3\) were calcined in air for 10 hours at 1100 Celsius, and then the powders were reduced to 0.5–1.0 μm diameter particles by ball milling. The slurry consisting of 60-70 wt% BaW ferrite fine powders and 30-40 wt% alcohol solution was cast into a mold, aligned by the application of an external magnetic field, and pressed into
pellets with a diameter of 4.0 mm and thickness of 0.5 mm under a pressure of 3.5 MPa. The magnetic field direction was parallel to the applied stress. Finally, the ferrite samples were sintered at 1020-1100°C for 4 h.

Figure 2-1: Flowchart of conventional ceramic process consisted of repeated steps of sintering and ball milling in this work.

The degree of crystallographic orientation was obtained from Philips X’pert PRO X-ray diffraction (XRD) measurements at room temperature and in a θ-2θ geometry using
a CuKα radiation source. The morphology was examined using a Hitachi S-4800 ultrahigh-resolution scanning electron microscope (SEM). Static magnetic properties were measured using a Lakeshore vibrating sample magnetometer (VSM) at room temperature.

2.2.2 Hexagonal crystal structure and morphology

Figure 2-2: X-ray diffraction patterns of BaZn$_{2-x}$Co$_x$Fe$_{16}$O$_{27}$ powders. Data collected at room temperature and in a θ-2θ geometry using a CuKα radiation source.

The XRD patterns of BaZn$_{2-x}$Co$_x$Fe$_{16}$O$_{27}$ (x=0.15, 0.20, and 0.25) powders are shown in Fig. 2-2. All diffraction features have been indexed to space group P63/mmc (published pattern: ICDD-#78-0135), corresponding to a hexagonal crystal structure.
Fig. 2-3 shows SEM images of BaW ferrite compacts, where Fig 2-3 (a), 2-3 (b), and 2-3 (c) present the morphology of the natural surface of BaZn$_{2-x}$Co$_x$Fe$_{16}$O$_{27}$ pellets with $x=0.15$, 0.20, and 0.25, respectively.
Figure 2-3: Scanning electron micrographs of $\text{BaZn}_{2-x}\text{Co}_x\text{Fe}_{16}\text{O}_{27}$ ferrites: (a) $x=0.15$ (surface), (b) $x=0.20$ (surface), (c) $x=0.25$ (surface), and (d) cross section of the compact.

It is observed that most of the grains have hexagonal structures and they are aligned along the c axis perpendicular to the sample plane. The grain sizes range from 0.4 to 1.0 $\mu$m and the average grain size is around 0.8 $\mu$m, which lies within the estimated single domain critical size range (0.5 $\sim$ 1.0 $\mu$m). It is of critical importance
that the grain sizes are well controlled in order to achieve high remanent magnetization. Fig 2-3 (d) is the cross section image of the aligned compact. It is seen that grains prefer to align along the hexagonal crystallographic c axis, perpendicular to the sample plane. A small misalignment is observed, which can be modified by optimizing the fabrication process.

2.2.3 Orientation and Magnetic Properties

The magnetic hysteresis loops were measured with the applied magnetic field aligned along the in-plane sample direction and perpendicular to the sample plane. Fig. 2-4 shows hysteresis curves in perpendicular direction for the 0.5 mm thick BaZn$_2$-$x$Co$_x$Fe$_{16}$O$_{27}$ ($x=0.15, 0.20, \text{and } 0.25$) pellets sintered at 1050 °C for 4 h. The detailed results are summarized in Table 2-1. The measured squareness ratio, $M_r/M_s$, were all above 70%, and the maximum was 79%, which demonstrate the discussion above based on SEM results that the grain size in the BaW ferrites has the scale of single domain size. Since Zn$_2$W ferrites have c-axis anisotropy, while Co$_2$W ferrites have planar anisotropy, doping Co can modify the anisotropy from c axis to c plane. It is noticed that the value of anisotropy field (Ha) decreased with Co substitution increased. The c-axis anisotropy was seen obviously in Fig. 2-4 (b), which matches with SEM results.
Figure 2-4: Hysteresis loops for BaZn$_{2-x}$Co$_x$Fe$_{16}$O$_{27}$ ferrites: (a) perpendicular direction with various x values, (b) in-plane and out-of-plane directions with x=0.20.
Table 2-1: Magnetic results for BaZn$_{2-x}$Co$_x$Fe$_{16}$O$_{27}$ ($x=0.15$, 0.20, and 0.25) samples.

<table>
<thead>
<tr>
<th>BaZn$<em>{2-x}$Co$<em>x$Fe$</em>{16}$O$</em>{27}$</th>
<th>$H_c$ (Oe)</th>
<th>$H_a$ (Oe)</th>
<th>$4\pi M_s$ (G)</th>
<th>$M_r/M_s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x=0.15$</td>
<td>2303</td>
<td>10600</td>
<td>2253</td>
<td>76%</td>
</tr>
<tr>
<td>$x=0.20$</td>
<td>2133</td>
<td>10000</td>
<td>2648</td>
<td>79%</td>
</tr>
<tr>
<td>$x=0.25$</td>
<td>2071</td>
<td>9600</td>
<td>2450</td>
<td>73%</td>
</tr>
</tbody>
</table>

2.2.4 Conclusion

In summary, we successfully prepared W-type barium hexaferrites BaZn$_{2-x}$Co$_x$Fe$_{16}$O$_{27}$ where $x=0.15$, 0.20, and 0.25. With Co ion doping, the anisotropy field is decreased while maintaining crystallographic c-axis texture. The XRD results confirmed a pure phase hexagonal crystal structure. The magnetic hysteresis loops revealed that the BaW ferrites as pressed pellets have apparent c-axis anisotropy and good squareness ($M_r/M_s \sim 79\%$), which was also confirmed by SEM results. The BaW hexaferrites have potentials for applications in low frequency self-biased microwave/millimeter devices, such as circulators and isolators.

2.3 Ni substituted W-type barium ferrites

Microwave circulators are one of the key elements in transmit and receive (T/R) modules in radar and communication systems. Most ferrite-based circulators are functioned under the strong biased field provided by an external magnet. The growing
needs of reducing size and cost in microwave integrated circuits simply presents a challenge in making novel low loss and self-biased ferrites, especially at operating frequencies below 20 GHz. The hexaferrites have adjustable magnetic anisotropy fields \((H_A)\) and ferromagnetic resonance (FMR) frequencies, so that they have been widely used in a variety of microwave ferrite devices. Although we have demonstrated the design of self-biased circulators at Ku-band based on self-biased M-type hexaferrites, high \(H_A\) and FMR frequency are still an obstacle in achieving circulators with low operating frequency and low insertion loss. In this work, self-biased W-type barium ferrite (BaW) was prepared and investigated on structure and magnetic properties.

### 2.3.1 Synthesis of Ni substituted W-type ferrites

Polycrystalline BaW ferrites having composition \(\text{BaNi}_{1.6}\text{Co}_{0.4}\text{Fe}_{16}\text{O}_{27}\) were prepared by a conventional ceramic process that entailed repeated steps of firing and ball milling. Starting materials of \(\text{BaCO}_3\), \(\text{NiO}\), \(\text{Co}_3\text{O}_4\), and \(\text{Fe}_2\text{O}_3\) were calcined in air for 10 hours at 1250°C, and then the powders were reduced to 0.5–1.0\(\mu\)m diameter particles by ball milling. The slurry consisting of 60-70 wt% BaW ferrite fine powders and 30-40 wt% alcohol solution was cast into a mold, aligned by the application of an external magnetic field, and pressed into pellets with a diameter of 4.6 mm and thickness of 0.2 mm under a pressure of 3.5 MPa. The magnetic field direction was parallel to the applied stress. Finally, the ferrite samples were sintered at 1020-1100°C for 4 h.

### 2.3.2 Structure and characterization

The XRD pattern of \(\text{BaNi}_{1.6}\text{Co}_{0.4}\text{Fe}_{16}\text{O}_{27}\) powders was shown in Fig. 2-5. All diffraction features have been indexed to space group P63/mmc (published pattern: ICDD-#78-0135), corresponding to a hexagonal crystal structure.
Figure 2-5: X-ray diffraction patterns of BaNi$_{1.6}$Co$_{0.4}$Fe$_{16}$O$_{27}$ powders.

Fig. 2-6 shows SEM images of BaW ferrite compacts at varies sintering temperature. It is observed that most of the grains have hexagonal structures and they are aligned along the c axis perpendicular to the sample plane. The grain sizes in Fig. 2-6 (a) range from 0.4 to 1.0 μm and the average grain size is around 0.8 μm, which lies within the estimated single domain critical size range (0.5 ~ 1.0 μm). In fig. 2-6 (b), it was seen the average grain size increased to 0.8~1.2 μm.
Figure 2-6: Scanning electron micrographs of BaNi$_{1.6}$Co$_{0.4}$Fe$_{16}$O$_{27}$ ferrites (natural surface) at varies sintering temperature: (a) T=1050 °C (b) T=1100 °C

2.3.3 Magnetic properties

The magnetic hysteresis loops were measured with the applied magnetic field aligned along the in-plane sample direction and perpendicular to the sample plane, which
was shown in Fig. 2-7. The detailed results are summarized in Table. 2-2. The measured squareness ratio, $M_r/M_s$, was achieved as high as 81%.

![Hysteresis loops for BaNi$_{1.6}$Co$_{0.4}$Fe$_{16}$O$_{27}$ ferrites at varies sintering temperature: (a) T=1050 °C, (b) T=1100 °C](image)

**Figure 2-7**: Hysteresis loops for BaNi$_{1.6}$Co$_{0.4}$Fe$_{16}$O$_{27}$ ferrites at varies sintering temperature: (a) T=1050 °C, (b) T=1100 °C.
Table 2-2: Magnetic results for BaNi$_{1.6}$Co$_{0.4}$Fe$_{16}$O$_{27}$ samples.

<table>
<thead>
<tr>
<th>Sintering $T$ (°C)</th>
<th>$M_s$ (emu/g)</th>
<th>$H_c$ (Oe)</th>
<th>$M_r/M_s$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1050</td>
<td>56</td>
<td>1754.9</td>
<td>81</td>
</tr>
<tr>
<td>1100</td>
<td>58</td>
<td>1216.6</td>
<td>72</td>
</tr>
</tbody>
</table>

2.3.4 Conclusion

In summary, we successfully prepared W-type barium hexaferrites BaNi$_{1.6}$Co$_{0.4}$Fe$_{16}$O$_{27}$. The XRD results confirmed a pure phase hexagonal crystal structure. The anisotropy field of this BaW ferrites decreased with the substitution of divalent Co ions, while, they maintained crystallographic c-axis texture. The measured anisotropy field was ~10 kOe. The magnetic hysteresis loops revealed that the BaW ferrites as pressed pellets have apparent c-axis anisotropy and good squareness ($M_r/M_s$~81%), which was also confirmed by SEM results. The BaW hexaferrites have potentials for applications in low frequency self-biased microwave/millimeter devices, such as circulators and isolators.

2.4 Reference


CHAPTER 3

LOW MAGNETIC LOSS U-TYPE HEXAFERRITE FILMS BY PULSED LASER DEPOSITION

3.1 Introduction to U-type hexaferrite materials

Hexagonal ferrites have the merits of low conductivity, moderate magnetic moment and high magnetocrystalline anisotropy, which make them of great interest and value in many microwave as well as permanent magnet applications [1-2]. A c-axis textured barium hexaferrite can result in low microwave loss and high hysteresis loop squareness required for the design of self-biased circulators [3-7] in which the external permanent magnet is removed from the circuit. It is crucial for low frequency microwave devices (1-20 GHz) to reduce the magnetic anisotropy field and in turn ferromagnetic resonance (FMR) frequency. In general, M-type, W-type and U-type hexaferrites all exhibit c-axis preferred magnetization, yielding magnetocrystalline anisotropy fields of ~17 kOe, ~12.7 kOe, and ~10 kOe, respectively. Concomitantly, the three structures demonstrated similar saturation magnetization, which makes U-type hexaferrite favorable to applications of X-band microwave devices due to lower anisotropy field. However, it is believed that ferrite films are of better compatibility with monolithic microwave integrated circuits (MMIC)
than bulk materials and for that reason we focus our attention here on the growth and refinement of the film system.

U-type barium hexaferrite has a chemical formula $\text{Ba}_6\text{Me}_2\text{Fe}_{36}\text{O}_{60}$, usually denoted as $\text{Me}_2\text{U}$, where $\text{Me}$ represents a divalent transition metal ion. It is a combination of M- and Y-type hexaferrites with crystal structure described by stacking sequence RSR*S*TS*.

Although U-type barium hexaferrite was found for several decades, the research on $\text{Me}_2\text{U}$ ferrite is much less than the other hexaferrite bulk materials [8-13]. Therefore, a study of U-type hexaferrite films enables one to not only develop fundamental research but also to explore novel planar devices.

3.2 Epitaxial growth of U-type hexaferrite films

In this work, we aim to focus on the epitaxial growth of U-type hexaferrite films grown by pulsed laser deposition (PLD) and their microwave properties. Co substituted Ni$_2$U hexaferrite films with c-axis texture and anisotropy field of ~8 kOe are studied here.

Growth conditions, crystallographic structure, magnetic properties and microwave performance are presented. This work reports for the first time on U-type hexaferrite films with low FMR linewidth, low magnetic anisotropy and high hysteresis loop squareness, which has great potential for applications of microwave devices operating at X-band.

3.2.1 Preparation of U-type hexaferrite films

Me$_2$U thin films were deposited on single crystal sapphire, (0001) Al$_2$O$_3$, substrates by a KrF excimer laser operating at a wavelength of 248 nm with energy per pulse of ~200 mJ. Fig. 3-1 shows the schematic of a PLD chamber. A homogeneous Me$_2$U target ($\text{Ba}_6\text{Ni}_{1.4}\text{Co}_{0.6}\text{Fe}_{36}\text{O}_{60}$) was made by the conventional ceramic process. A calculation
indicates that the lattice mismatch between the c-plane oriented sapphire substrate and Me$_2$U ferrite film is ~7%, whereas the difference in thermal expansion coefficient is up to ~20% over the range of 0 – 920°C. The depositing pressure and substrate temperature were held constant at an oxygen pressure of 50 mTorr and a temperature of 900 °C, respectively. The laser pulse repetition rate was increased from 1 to 10 Hz for the initial 10 min of a deposition in order to enhance the adhesion at the interface, and then fixed at 10 Hz for the remaining time. The post-deposition anneal was performed in a tube furnace at 1150°C for 20 min in air. The films have a thickness ranging from 100-200 nm measured by a scanning surface profilometer.

![Schematic of a PLD chamber](image)

**Figure 3-1:** Schematic of a PLD chamber.
Crystallographic structure and orientation were determined by Philips X’pert PRO X-ray diffraction (XRD) measurements at room temperature in a θ-2θ geometry using a CuKα radiation source. The morphology was examined using a Hitachi S-4800 ultrahigh-resolution scanning electron microscope (SEM). Static magnetic properties were measured using a Lakeshore vibrating sample magnetometer (VSM) at room temperature. At 9.65 GHz (X-band), the FMR linewidth of the samples was measured by using a field sweep electron paramagnetic resonance spectroscopy (EPR).

3.2.2 Crystallographic structure and surface morphology

![X-ray diffraction pattern](image)

**Figure 3-2:** X-ray diffraction patterns of Me₂U hexaferrite thin films. Data was collected in a θ-2θ geometry using a CuKα radiation source, and a logarithmic scale is used for intensity (y-axis).
The crystal structure of annealed Me₂U ferrite films were characterized by XRD and the patterns are shown in Fig. 3-2. All of the diffraction lines presented were indexed to U-type hexaferrite crystallographic structure with a rhombohedral space group R3m, indicating unit cell parameters \( a=5.88 \) Å and \( c=113.2 \) Å. However, most of the diffraction lines were indexed to the \((0 \ 0 \ n)\), \((1 \ 0 \ n)\), and \((0 \ 1 \ n)\) planes. It is worth noting that either \((1 \ 0 \ n)\) or \((0 \ 1 \ n)\) planes only deviate by \(\sim 0.2^\circ\) from the \(c\)-axis, which may still give rise to high magnetic anisotropy out of the film plane. It is evident that the annealed films have a single phase crystal structure and a strong \(c\)-axis preferred orientation perpendicular to the film plane.

![Pole figure measured along the (0 0 39) diffraction peak.](image)

**Figure 3-3:** Pole figure measured along the \((0 \ 0 \ 39)\) diffraction peak.
In order to characterize the crystal structure of the prepared Me$_2$U thin film within c-plane, a pole figure measurement is implemented along (0 0 39) diffraction peak. The angle between the film normal and the vector bisecting the incident and detected x-ray beams, φ, was varied from 0° to 90°. For each φ, the scanning runs along the azimuthal angle about the bisecting vector, ξ, varied from -100° to 200°. For a discrete scanning with φ from 0° to 90°, only around φ =30° the reflections shown a series of peaks when ξ varied from -100° to 200°. Fig. 3-3 shown the data collecting at φ =30° from peak (0 0 39). It is seen that five peaks shown at ξ equals to -64°, -4.5°, 56°, 116.5° and 176.5°, respectively, with the range from -100° to 200°. The distances between every adjacent two peaks are Me$_2$U thin films. It is observed that all of the grains have triangular shapes with sharp edges. They are aligned along the c-axis perpendicular to the sample plane. Since U-type hexaferrites belong to rhombohedral space group, six formula units form a triple-primitive hexagonal unit cell. Thus each triangle in Fig.2 represents one formula unit (Ba$_2$MeFe$_{18}$O$_{30}$), and several rhombus composed of two triangles can be seen in the figure. It is believed that this surface image shows an intermediate stage of the epitaxial growth of Me$_2$U films.
3.2.3 Static magnetic properties of U-type ferrite films

The magnetic hysteresis loops were measured with an applied magnetic field along the film plane and perpendicular to the film plane. Fig. 3-5 (a) shows hysteresis curves of as-deposited Me$_2$U thin films, while Fig. 3-5 (b) and (c) show the results of films annealed at 1050°C and 1100°C for 20 min. in air, respectively. Obviously, the annealing improves c-axis orientation, which is consistent with the XRD results. Meanwhile, a squareness ratio, $M_r/M_s$, increases from 65% to 92% upon annealing at 1100°C. Additionally, the annealed films exhibit a saturation magnetization ($4\pi M_s$) of ~3.6 kOe, and a remanent magnetization of ~3.3 kOe. Furthermore, a magnetic anisotropy field was determined from Fig. 3-5 (c), $H_k$ to be ~8 kOe. This is in good agreement with our expectation in that Co ion decreases magnetic anisotropy fields. Generally a Co$_2$U barium hexaferrite gives rise to c plane anisotropy [14].
Figure 3-5: Hysteresis loops for Me$_2$U thin films: (a) before annealing, (b) after annealing at 1050°C for 20min, and (c) annealing at 1100°C.
3.2.4 Microwave magnetic properties of U-type ferrite films

Figure 3-6: Variation of ferromagnetic resonance absorption derivative with field at $f=9.65$ GHz for Me$_2$U thin films.

The FMR measurements were carried out using a microwave cavity at X-band ($f=9.65$ GHz) with a static magnetic field applied parallel to the film plane. The resonance condition in this configuration is given by

$$f = \gamma' \sqrt{(H_k - 4\pi M)^2 - H^2}$$

where $f$ is the FMR frequency, $\gamma'$ is the gyromagnetic ratio over $2\pi$, $H_k$ is the uniaxial magnetic anisotropy field, $4\pi M$ is magnetization, and $H$ is the externally applied magnetic field. Figure 3-6 shows the differential power absorption $dP/dH$ versus $H$, and this absorption curve shows an FMR linewidth of 200 Oe, which is comparable with the
linewidth (210 Oe) as reported in bulk Me$_2$U ferrite single crystal [12]. The uniaxial anisotropy field for Me$_2$U films was calculated to be $H_k=8.2$ kOe from Eqn. (3-1), which is very close to the one derived from magnetization measurements depicted in Fig.3-5 (c).

3.2.5 Temperature dependence of magnetic properties

![Graph showing temperature dependence of magnetization under various applied magnetic field for U-type thin films.]

**Figure 3-7:** Temperature dependence of magnetization under various applied magnetic field for U-type thin films.

The temperature dependence of the magnetization for U-type thin films was measured between 300 K and 900 K under various applied magnetic field along the c-plane of the
samples, is shown in Fig. 3-7. It is observed that the Curie temperature of prepared U-type thin film is around 700k, which matches with data in refs [15]. It is clear that at applied field= 100 Oe, the magnetization curve shows a peak around 450 K, which is the evidence of phase transition under low applied external field. It is studied by K. Okumura et al, that U-type is a room-temperature magnetoelectric material who share the same mechanism with other magnetoelectric materials, like Z or Y-type hexaferrites [16]. It is speculated in the prepared U-type film there is a phase transition from conical order to ferrimagnetic structure at 450 K.

### 3.3 Conclusion

The characterization and growth of U-type barium hexaferrite thin films deposited on basal plane oriented sapphire substrates by pulsed laser deposition were systematically investigated. The XRD results confirmed a pure phase rhombohedral crystal structure observed having a strong c-axis crystallographic texture. The measurements are in accordance with the observation of VSM and SEM. By means of proper post-deposition thermal treatments, a high magnetic squareness ($M_r/M_s=92\%$) was achieved. The uniaxial magnetocrystalline anisotropy field was determined to be $\sim8$ kOe. Importantly, the FMR linewidth was measured to be 200 Oe at X-band frequency. These results reveal that the Me$_2$U hexaferrite films are good candidates for low frequency self-biased microwave/millimeter device applications.

### 3.4 Reference


dependence of Ba₄Zn₂–xCoₓFe₃₆O₆₀ U-type hexaferrites prepared by citrate sol–gel on
166.


Kimura, “Magnetism and magnetoelectricity of a U-type hexaferrite Sr₄Co₂Fe₃₆O₆₀”, Appl.
CHAPTER 4

LOW LOSS CO$_2$Z FERRITE COMPOSITES
WITH EQUIVALENT PERMITTIVITY AND PERMEABILITY

4.1 Introduction to Z-type ferrites

Improved performance and miniaturization are needed to meet the ever-increasing demands of devices used in ultra-high frequency (UHF), L-band, and S-band, which are of particular interest in a variety of commercial and defense related applications [1-8]. Utilizing materials possessing high permeability and permittivity with low magnetic losses is a promising solution. As a critical component in radar and modern wireless communication systems, antenna elements with compact size are constantly sought [9-10]. The use of magnetodielectric substrates with values of $\varepsilon'$ identical to $\mu'$ is popular in designing two types of RF devices: electromagnetic band-gap (EBG) structures and miniaturized antennas [11]. The magneto-dielectric materials can miniaturize both the EBG structures and antenna by the same factor however using moderate values of permittivity and permeability ($n = \sqrt{\varepsilon'\mu'}$) compared to the dielectric-only high permittivity material. The use of $\varepsilon'$ identical to $\mu'$ has two benefits: for multilayer EBG structures, it produces better band-gap rejection levels [11]; for antennas, It’s either low $\varepsilon'$ or high $\mu'$ is responsible for the broadening of patch antenna bandwidth [12], while An equivalent $\varepsilon'$ to
µ’ allows for ease of impedance matching over a much wider bandwidth, which gives more flexibility of antenna design in order to simply fabrication and efficiency of antennas. However, it has been extremely challenging to realize such materials for use at UHF. Previous results with these same aims produced materials exhibiting relatively high magnetic loss 0.45 at 0.8 GHz [10]. Compared to spinel ferrites having high permeability and low cut-off frequency, some hexaferrites, such as cobalt-substituted barium Y-type (Co₂Y) and Z-type (Co₂Z), have much higher ferromagnetic resonance frequencies and permeabilities [13-15]. These outstanding magnetic properties make hexagonal ferrites attractive in high frequency applications, e.g. inductors, filters and antennas. Many studies on microwave properties of Z-type hexaferrites, represented by complex permittivity and permeability, have been reported [16-20].

4.2 Co₂Z ferrite composites

In this work, a series of iridium substituted Z-type barium ferrites (Co₂Z) were prepared and studied for their structure and microwave properties. It was revealed that the Y-type phase appears as a secondary phase with the doping of iridium, which helps to dramatically reduce the dielectric and magnetic losses. Effective medium approximations were adopted here to analysis this duel phase material. Bi₂O₃ was introduced into the hexaferrite materials to match the permittivity and permeability. It was shown almost equal values of ε’ and µ’ in the UHF band were achieved, and also with the low dielectric and magnetic losses, which makes the composite ferrite a desirable candidate as microwave magnetodielectric substrate materials.
4.2.1 Preparation of iridium substituted Co$_2$Z ferrite composites

Polycrystalline Co$_2$Z ferrites, having nominal composition Ba$_3$Co$_{2+x}$Ir$_x$Fe$_{24-2x}$O$_{41}$, where x=0, 0.05, 0.10, 0.15 and 0.20, were prepared by a two-step conventional ceramic process. Starting materials of BaCO$_3$, IrO$_2$, Co$_3$O$_4$, and Fe$_2$O$_3$ were calcined in air for 6 hours at 1000°C, and then crushed and ball milled. The mixture, consisting of 90 vol-% ferrite fine powders and 10 vol-% polyvinyl alcohol (PVA) binder, was pressed into a toroid with an outer diameter of 7 mm, inner diameter of 3 mm, and width of about 2 mm. This sample size is adequate for microwave measurements. Since oxygen atmosphere can help decrease the dielectric loss, the ferrite samples were sintered at 1250-1280 ºC for 4 h in flowing oxygen gas as a final process step.

Crystallographic structure was determined by X-ray diffraction (XRD, Philips X’pert PRO) measurements at room temperature in a θ-2θ geometry using CuKα radiation. The complex permittivity and permeability spectra were measured over a frequency range from 0.3–1 GHz with an Agilent E864A 45MHz-50GHz PNA series network analyzer and a 7 mm HP 85050C precision airline. In this letter, we will use permeability and permittivity values, as the relative permeability and the relative permittivity, respectively.

4.2.2 Crystal structure of composite ferrite samples

The crystal structure of composite ferrite samples was characterized by XRD and the patterns are shown in Fig. 4-1. All of the diffraction lines were indexed to either Z-type or Y-type (marked by red dot in Fig. 1) hexaferrite crystallographic phases. Generally, it is inevitable to synthesize bulk Z-type ferrite materials without impurity phases of W- and/or Y-type. The Z-type phase is the dominant phase when heat-treated at temperatures higher than 1200°C In the range of 1200-1300°C, the impurity is the Y-type phase, and when higher
than 1300 °C, it is the W-type phase. It is seen in Fig. 1, for x=0, the results show an almost pure Z-type phase with a small amount of Y-type phase. With an increase in x, not only does the appearance of new Y-type peaks increase, but also the peak intensities can be clearly seen to increase. The amount of Z-type and Y-type phases was calculated based on XRD data and the percentages are shown in Fig. 4-2. The weight percentage of Z-type phase deceased from 97.5% to 65.1% with x values increasing to 0.2. It is obvious that doping with iridium changes the crystal structure from a quasi-single phase to a two phase system.

![X-ray diffraction patterns](image)

**Figure 4-1:** X-ray diffraction patterns of composite ferrite samples with various x. Data were collected at room temperature in a θ-2θ geometry using a CuKα radiation source.
4.2.3 **Effective medium approximation**

In order to investigate the affect upon the microwave properties of the dual phase composition of the ferrite composites, an effective medium approximation was used. The most common theories are Maxwell-Garnet (MG) and Bruggeman equations [21-22]. The MG model is generally expected to be valid for small or high filling factors, while in the Bruggeman model the two phases contribute equally. Thus the Bruggeman model is often used for moderate filling factors. In our case, the Z-type phase is dominant and thus the MG approximation was applied. It was assumed that the composite ferrites comprised of spherical inclusions with random distribution, the effective permittivity $\varepsilon_e$ and permeability $\mu_e$ are given by:
\[\varepsilon_e = \varepsilon_Y + 3p\varepsilon_Y \frac{\varepsilon_Z - \varepsilon_Y}{\varepsilon_Z + 2\varepsilon_Y - p(\varepsilon_Z - \varepsilon_Y)} \quad (4 - 1)\]

\[\mu_e = \mu_Y + 3p\mu_Y \frac{\mu_Z - \mu_Y}{\mu_Z + 2\mu_Y - p(\mu_Z - \mu_Y)} \quad (4 - 2)\]

, where \(\varepsilon_Z, \varepsilon_Y, \mu_Y\) and \(\mu_Z\) are the permittivity and permeability of Z-type and Y-type phases, respectively, and \(p\) is the volume fraction of Z-type phase. The real part of the effective permittivity and permeability of the composite ferrites with variation of volume fraction is shown in Fig. 4-3. The symbols represent the experimental data and the dash lines are the best fitted results from applying Eqns. (4-1) and (4-2). Since Y-type and Z-type ferrites possess nearly the same bulk density \(\sim 5.3\) g/cm\(^3\) [23], the calculated phase percentage in weight can be easily converted into a volume fraction, which is the x-axis of Fig. 4-3. The relationship between experiment and theory are in good agreement using the MG approximation. In the fitting procedures, the values of \(\varepsilon_Z, \varepsilon_Y, \mu_Y\) and \(\mu_Z\) should be identified first - the fitting can then be determined by applying Eqns. (1) and (2). The values of \(\varepsilon_Z\) and \(\mu_Z\) of pure Co\(_2\)Z ferrites are readily available from our previous work [24]. However, identifying values of \(\varepsilon_Y\) and \(\mu_Y\) are more challenging to determine since the precise formula of the Y-type phase in this composite is unknown. Here we choose values from ref. [23, 25] due to similar elemental composition, process history, grain size, etc. A deviation with effective permittivity in Fig. 4-3 is observed when the volume fraction of the Z-type phase deceases to 70%, this may be due to the above explanations. Also a discrepancy exists in the effective permeability seen in the middle range of the experimental data. Besides the above explanation, another possible reason is that the frequency dispersion law will affect the effective medium approximation. For the ferrite materials with single-domain ellipsoidal particles and a uniaxial anisotropy, the frequency-dependent permeability obeys the Lorentzian dispersion law [26]. In reality, the dispersion curve of a polycrystalline
sample will deform due to the inhomogeneous microstructure or domain-wall dynamics. Only if the inclusions of the composite have Lorentzian dispersion shape will the effective dispersion curve derived from the MGA mixing rule be Lorentzian as well, otherwise it will be distorted.

![Graph](image)

**Figure 4-3:** Real permittivity or permeability with various Co$_2$Z volume fractions.

Values of real permittivity and real permeability of composite ferrites affected by doping with iridium at 0.8 GHz are shown in Fig. 4-4. The four curves in Fig. 4-4 (a) represent real permittivity, real permeability, dielectric loss and magnetic loss. For lightly doped samples, the four parameters deceased significantly. With further increases in iridium content, these parameters decrease more slowly. The values of loss tan $\delta_e$ and loss tan $\delta_\mu$ attain their minimum at $x =0.12$ and then increase, which are clearly seen in Fig. 4-
4 (b). The change in parameters with various x in percentage is shown in Fig. 4-4 (b). The loss tan δ_ε and loss tan δ_µ values decreased 80% and 90%, for x = 0.12 and 0.15, respectively, while the real permittivity and real permeability decreased 30% and 50%, respectively. The decrease of dielectric and magnetic loss is nearly twice as much as that of the real permittivity and permeability.

**Figure 4-4:** Permittivity and permeability of composite ferrites with various x at 0.8 GHz.
Figure 4-5: The real permittivity and real permeability for sample A, B and C over 0.3-1 GHz.

Next, the effect of the additive Bi₂O₃ upon the complex permittivity and permeability of the composite ferrites was examined. Samples labeled A, B and C with various amounts of Bi₂O₃ and iridium content (i.e., x=0.12-0.15) were prepared for microwave measurements. Figure 4-5 shows the permittivity and permeability spectrum of the three samples in the frequency range from 0.3 GHz to 1 GHz. The values of real permittivity are very close to those of the real permeability for all three samples. Sample
C however shows the best performance in impedance matching to free space with equivalent values of real permittivity and permeability. As for the dielectric and magnetic loss, all three samples have relatively lower loss tan δε and loss tan δµ at frequencies from 0.5 GHz to 0.9 GHz compared with data of the barium Z-type ferrites reported in reference 14-19. Sample A shows lower magnetic loss than samples B and C in the high frequency range of 0.6-1.0 GHz. The detailed results of permittivity, permeability, loss tan δε and loss tan δµ at five different frequencies are summarized in Table. 4-1. It was found that the values of μ’/ε’ are equal to 1.0 over the frequency range from 0.65 GHz to 0.85 GHz for sample C, which means the characteristic impedance is the same as that of free space. The loss tan δε of all the samples over the whole frequency range remains low at around 0.07. The loss tan δµ increases from 0.17 to 0.29 with frequencies from 0.65 GHz to 0.85 GHz. This is the result of the frequency approaching that of the FMR frequency of the samples. Furthermore, the loss factors (tanδε/ε’ and tanδµ/µ’) were calculated to be 0.008 and 0.037 at 0.8 GHz, respectively, in order to make comprehensive performance evaluation of the ferrite materials. As shown above, it is impressive that both magnetic loss (0.06 at 0.8 GHz) and loss factor (0.27 at 0.8 GHz) of the present ferrite composites represent the lowest values among those reported for ferrites at UHF.

**Table 4-1**: ε’, μ’, tan δε and loss tan δµ of sample A, B and C at five different frequencies

<table>
<thead>
<tr>
<th>Sample name</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.65GHz</td>
<td>ε’</td>
<td>6.48</td>
<td>8.01</td>
</tr>
<tr>
<td>Frequency</td>
<td>$\mu'$</td>
<td>$\varepsilon'$</td>
<td>$\tan\delta_\varepsilon$</td>
</tr>
<tr>
<td>-----------</td>
<td>--------</td>
<td>---------------</td>
<td>-------------------</td>
</tr>
<tr>
<td>0.70GHz</td>
<td>4.70</td>
<td>6.52</td>
<td>0.05</td>
</tr>
<tr>
<td></td>
<td>4.69</td>
<td>8.12</td>
<td>0.05</td>
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<tr>
<td></td>
<td>4.69</td>
<td>7.49</td>
<td>0.05</td>
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<td>0.08</td>
</tr>
<tr>
<td></td>
<td>4.70</td>
<td>7.49</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>4.70</td>
<td>7.49</td>
<td>0.08</td>
</tr>
<tr>
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</tr>
<tr>
<td></td>
<td>4.70</td>
<td>7.44</td>
<td>0.13</td>
</tr>
<tr>
<td></td>
<td>4.70</td>
<td>7.44</td>
<td>0.13</td>
</tr>
<tr>
<td></td>
<td>$\tan\delta_\varepsilon$</td>
<td>$\tan\delta_\mu$</td>
<td>$\tan\delta_\varepsilon / \varepsilon'$</td>
</tr>
<tr>
<td>----------------</td>
<td>--------------------------</td>
<td>------------------</td>
<td>------------------------------------------</td>
</tr>
<tr>
<td>$\tan\delta_\varepsilon$</td>
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<td>0.07</td>
<td>0.06</td>
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<tr>
<td>$\tan\delta_\mu$</td>
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<td>0.25</td>
<td>0.27</td>
</tr>
<tr>
<td>$\tan\delta_\varepsilon / \varepsilon'$</td>
<td>0.011</td>
<td>0.009</td>
<td>0.008</td>
</tr>
<tr>
<td>$\tan\delta_\mu / \mu'$</td>
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<td>0.033</td>
<td>0.037</td>
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<table>
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<th>$\varepsilon'$</th>
<th>$\mu'$</th>
<th>$\tan\delta_\varepsilon$</th>
<th>$\tan\delta_\mu$</th>
<th>$\tan\delta_\varepsilon / \varepsilon'$</th>
<th>$\tan\delta_\mu / \mu'$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon'$</td>
<td>6.58</td>
<td>8.16</td>
<td>7.29</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>$\mu'$</td>
<td>4.64</td>
<td>7.37</td>
<td>7.22</td>
<td></td>
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<td></td>
</tr>
<tr>
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<td>0.07</td>
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<tr>
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<td>0.29</td>
<td></td>
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<td></td>
</tr>
<tr>
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<tr>
<td>$\tan\delta_\mu / \mu'$</td>
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<td>0.037</td>
<td>0.040</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Sample A

[Image of SEM micrograph with a scale bar of 100 μm]
The Scanning electron micrographs of sample A, B and C are shown in Fig. 4-6. It is observed that the particle size in sample B is around 100-200 μm, while the particle size in sample C is around 100-150 μm.
Figure 4-7 shows the measured microwave properties of sample C in the frequency range of 0.55-0.85 GHz. The normalized impedance of the prepared composite hexaferrites was around 1, which achieved the goal of impedance matching to free space. The miniaturization factor, which is also known as the refractive index, was 7.3 over the frequency range. The measured results proved the prepared composite hexaferrites to be promising magnetodielectric materials for antenna substrate.

**Figure 4-7:** Measured microwave properties of sample C in the frequency range of 0.55-0.85 GHz.
4.3 Conclusion

In summary, polycrystalline ferrite composites of nominal composition \( \text{Ba}_3\text{Co}_{2+x}\text{Ir}_x\text{Fe}_{24-2x}\text{O}_{41} \), where \( x = 0, 0.05, 0.10, 0.15 \) and 0.20, were prepared by conventional ceramic processes. XRD studies revealed that the samples ranged from a nearly pure Z-type phase to composites with substantial Y type as a secondary phase. Importantly, the second phase (Y-type) can effectively tailor microwave dielectric and magnetic properties, showing obvious reduction in the loss tan \( \delta_e \) and loss tan \( \delta_\mu \) by 80% and 90% at 0.8 GHz with the addition of iridium having \( x = 0.12-0.15 \), respectively. Additionally, \( \text{Bi}_2\text{O}_3 \) can balance the real permittivity and permeability over the frequency range of 0.3 GHz to 1 GHz to make the characteristic impedance the same as that of the free space impedance. The loss factors (\( \text{tan}\delta_e/\varepsilon' \) and \( \text{tan}\delta_\mu/\mu' \)) were the lowest values for randomly-oriented polycrystalline ferrite samples among those reported previously. These properties make these composite ferrites suitable for applications in microwave devices at ultra-high frequency (UHF), such as novel miniature antennas.

4.4 Reference


CHAPTER 5

TUNABLE PERMITTIVITY AND PERMEABILITY OF LOW LOSS Z+Y-TYPE FERRITE COMPOSITES

5.1 Introduction to low loss ferrites

In modern wireless communication systems the demand for high performance devices of miniaturized structure is ever increasing [1-9]. High permeability with equivalent permittivity having low magnetic/dielectric losses are ideal as antenna substrate materials. Such materials enable miniaturization, while concomitantly maintaining or enhancing antenna gain and bandwidth. Spinel ferrites, with high permeability values, such as NiZn ferrites, have been widely used for high frequency applications. However, these ferrites exhibit relatively low cutoff frequencies that prevent their use above 0.3 GHz. Some hexaferrite materials, such as cobalt substituted barium Y-type (Co$_2$Y) and Z-type (Co$_2$Z) have much higher ferromagnetic resonance frequencies of above 1.0 GHz due to their high magnetocrystalline anisotropy fields and high permeabilities [10-12]. Studies on microwave properties of Y-type or Z-type hexaferrites, represented by complex permittivity and permeability, have been reported [13-16]. Accordingly, the use of magnetodielectric substrates with values of $\varepsilon'$ equivalent to $\mu'$ is popular due to the ease in impedance matching between substrate and free space ($Z = Z_0(\sqrt{\mu'/\varepsilon'})$, where $Z_0$ is
impedance of free space) among other benefits [17]. It turns to have free impedance matching if the materials used for antenna have same magnetic permeability ($\mu'$) and permittivity ($\varepsilon'$) over a much wider bandwidth. As a result, the use of $\varepsilon'$ equal to $\mu'$ gives more flexibility to antenna designers to simply fabrication and enhanced efficiency. It has been reported that Co$_2$Y has $\varepsilon'$ of 15-20 and $\mu'$ of 2-3, while Co$_2$Z has $\varepsilon'$ of 12 and $\mu'$ of 18-19 [18]. We therefore propose that it is possible to realize equivalent permittivity and permeability if one composite system consisting of the two hexaferrites is considered.

### 5.2 Z+Y-type ferrite composites

In particular, our previous work indicated [19] that the appearance of the Y-type ferrite phase in iridium doped Co$_2$Z ferrites not only dramatically reduces the dielectric and magnetic losses, but also allows tailoring of permittivity and permeability values. In the previous experiment, however, a phase fraction was tuned by the doping of iridium, which introduces serious cost restraints for commercial use. Therefore, we propose in this work that the ferrite composites be achieved by the mixing and sintering Co$_2$Y and Co$_2$Z ferrite particles without the substitution of iridium. A desirable equivalent permeability or permittivity is predicted by the effective medium approximation. We aim to make the ferrite composites of tunable permeability or permittivity and low dielectric and magnetic losses, as well as cost-effective for the mass production of microwave antenna substrates.

#### 5.2.1 Preparation of Z+Y-type ferrite composites

Polycrystalline Co$_2$Z and Co$_2$Y ferrite particles, having nominal composition Ba$_3$Co$_2$Fe$_{24}$O$_{41}$ and Ba$_2$Co$_2$Fe$_{12}$O$_{22}$, respectively, were prepared by a two-step conventional ceramic process. Starting materials of BaCO$_3$, Co$_3$O$_4$, and Fe$_2$O$_3$ were calcined in air for 6 hours at 1000°C. The crushed powers were mixed leading to a decreasing mass ratio of Z-
type ferrite from 100 wt% to 0 wt% and then dried after 4 hrs of wet-milling. These samples will henceforth be designated as Z100, Z75, Z50, Z25, and Z0. The mixed powders with 10 wt% polyvinyl alcohol (PVA) binder were pressed into a toroid with an outer diameter (OD) of 7 mm, inner diameter (ID) of 3 mm, and thickness of about 2 mm. The ferrite samples were sintered at 1240 °C for 4 h in flowing oxygen gas in order to reduce the magnetic or dielectric losses.

Crystallographic structure was determined by X-ray diffraction (XRD, Philips X’pert PRO) measurements at room temperature in a 0-2θ geometry using Cu-Kα radiation. An Agilent Vector Network Analyzer E5071C was used to measure the complex permittivity and permeability spectra over a frequency range from 0.1–10 GHz. All of the toroids for the frequency spectrum measurements were polished in the OD and ID to effectively match to the APC-7 test fixture. In this article, the permeability or permittivity represents the relative permeability or permittivity.

5.2.2 Crystal structure of Z+Y-type ferrite composites

The XRD patterns of composite ferrite sample Z100, Z75, Z50, Z25, and Z0 were characterized using a Cu-Kα source, as shown in Fig. 5-1. All of the diffraction lines were indexed to either Z-type or Y-type (marked by red dot in Fig. 5-1) hexaferrite crystallographic phases. It is seen in Fig. 5-1, the sample Z100 and Z0 show pure Z-type phase and Y-type phase, respectively. The other samples reveal a combination of Y-type and Z type hexaferrite phases. It is noticed that the intensity of the peaks indexed to the Z-type ferrite decreased with the reduction in fraction of Z-type ferrite in the composite, as expected.
**Figure 5-1:** X-ray diffraction patterns of composite ferrite sample Z100, Z75, Z50, Z25, and Z0. Data were collected at room temperature in a θ–2θ geometry using a Cu-Kα radiation source.

The Scanning electron micrographs of Z100, Z75, Z50, and Z25 are shown in Fig. 5-2. It is observed that the particle size in all these samples is around 5-10 μm. Z50 and Z25 are seen to have more fine grains than the other two samples.
5.2.3 Effective medium approximation

An effective medium approximation was used to predict the microwave properties of the Z-type and Y-type ferrite composites. In the most commonly used Maxwell-Garnet (MG) model [20-21], it is assumed that the composite ferrites comprise of spherical inclusions with random distribution, and no interactions between particles. The effective permittivity $\varepsilon_e$ and permeability $\mu_e$ are given by:
\[
\varepsilon_e = \varepsilon_Z + 3(1-p)\varepsilon_Z \frac{\varepsilon_Y - \varepsilon_Z}{\varepsilon_Y + 2\varepsilon_Z - (1-p)(\varepsilon_Y - \varepsilon_Z)} \tag{5-1}
\]

\[
\mu_e = \mu_Y + 3p\mu_Y \frac{\mu_Z - \mu_Y}{\mu_Z + 2\mu_Y - p(\mu_Z - \mu_Y)} \tag{5-2}
\]

, where \( \varepsilon_Z, \varepsilon_Y, \mu_Y \) and \( \mu_Z \) are the permittivity and permeability of Z-type and Y-type phases, respectively, and \( p \) is the volume percent of Z-type phase. The real part of the effective permittivity and permeability of the composite ferrites with variation in volume percent is depicted in Fig. 5-2. The experimental and calculated data are denoted as symbols and dash line curves, respectively. The x-axis in Fig. 5-2 represents the volume percent, which is the same as mass percent of the samples because Y-type and Z-type ferrites possess nearly the same bulk density \( \sim 5.3 \text{ g/cm}^3 \) [18]. As mentioned early, magnetodielectric materials with matched values of \( \varepsilon' \) and \( \mu' \) are favorable for use as substrate materials of miniature antenna due to the advantage of impedance matching between substrate and free space. It is predicted by theory (see Fig. 5-2) that the ferrite composite containing 90 vol\% Z-type will give rise to identical values of \( \varepsilon' \) and \( \mu' \), whereas the experimental data indicates closest values of \( \varepsilon' \) to \( \mu' \) to coincide with the 75 vol\% of Z-type ferrite component. This small discrepancy may be attributed to the effect of particle size and morphology upon permittivity, which is more visible than that on the permeability in this case. Nevertheless, the prediction of permeability with phase percent is in good agreement with the experimental data. In contrast, the measured permittivity for all of the composites is lower than calculated values.
5.2.4 Microwave properties of Z+Y-type ferrite composites

The permeability and permittivity spectra of samples Z100, Z75, Z50, Z25, and Z0 over a frequency range from 0.1 GHz to 10 GHz are shown in Figs. 5-3 and 5-4, respectively. In Fig. 5-3, the real permeabilities remain nearly constant at low frequencies, whereas the imaginary permeabilities show a distinct peak at cutoff frequencies. It is obvious that the cutoff frequency increases with the increased percent of Y-type ferrite phase that is attributed to the high magnetocrystalline anisotropy field (~28 kOe) of Co2Y ferrite. Correspondingly, the real permeability decreases with a reduction in Co2Z phase. In Fig. 5-4, both the real and imaginary permittivity of the composite samples decreases...
with increasing frequency. The dielectric loss tangent for Z100, Z75, Z50, Z25 and Z0 are 0.20, 0.04, 0.14, 0.36 and 0.41 at 0.65 GHz, corresponding to the magnetic loss tangent of 0.59, 0.25, 0.20, 0.17 and 0.17, respectively. Due to low magnetic/dielectric loss and high permeability, the Sample Z75 reveals the best performance among those samples. It has been verified that the Z-type and Y-type ferrite composites are promising magnetodielectric materials in which both $\varepsilon'$ and $\mu'$ are readily tunable. More importantly, the microwave magnetic and dielectric properties of sample Z75 are measured and depicted in Fig. 5-5. The values of real permittivity are nearly equivalent to real permeability over the frequency range from 0.1 to 1 GHz. It is also calculated for the sample Z75 that the ratio ($\mu'/\varepsilon'$) is equal to 0.91 ± 0.01 at frequencies from 0.55 to 0.75 GHz, while retaining a high $\mu'$ and $\varepsilon'$ ~12-13. Meanwhile, the dielectric loss ($\tan\delta_\varepsilon$) is of the order of magnitude of $10^{-2}$ over a frequency range from 0.1 to 1 GHz, whereas the magnetic loss is ($\tan\delta_\mu$) is below 0.29 for frequencies below 0.7 GHz. As a result, the values of $\varepsilon'$, $\mu'$, $\tan\delta_\varepsilon$ and $\tan\delta_\mu$ at 0.65 GHz are 13.5, 12.3, 0.04 and 0.25, respectively. Furthermore, the loss factors ($\tan\delta_\varepsilon/\varepsilon'$ and $\tan\delta_\mu/\mu'$) were calculated to be 0.003 and 0.02 at 0.65 GHz, respectively, allowing for a comprehensive performance evaluation of the ferrite materials to be made.
Figure 5-4: Permeability spectrum of Sample Z100, Z75, Z50, Z25, and Z0 over a frequency range of 0.1-10 GHz.
Figure 5-5: Permittivity spectrum of Sample Z100, Z75, Z50, Z25, and Z0 over a frequency range of 0.1-10GHz.
Figure 5-6: Microwave magnetic and dielectric properties of sample Z75 over 0.1-1 GHz.

5.3 Conclusion

In summary, a series of Z-type and Y-type polycrystalline ferrite composite samples, designated Z100, Z75, Z50, Z25, and Z0 with various phase concentrations, were prepared by conventional ceramic processes. The effective permittivity $\varepsilon_e$ and permeability $\mu_e$ were calculated in terms of Maxwell-Garnet approximation. The permeability and permittivity spectra of the ferrite composites were measured over a frequency range from 0.1 to 10 GHz. The experimental data exhibits a tunable permeability and permittivity with change in phase percent. In addition, the microwave magnetic and dielectric properties are
characterized and discussed. In particular, the composite with 75 vol% of Z-type ferrite leads to a value of permittivity nearly equivalent to permeability at frequencies from 0.1 to 1 GHz, while the dielectric and magnetic losses are less than 0.08 at $f < 1$ GHz and 0.29 at $f < 0.7$ GHz. Furthermore, Z75 shows particular value at 0.65 GHz with values of $\varepsilon'$, $\mu'$, $\tan\delta_{\varepsilon}$ and $\tan\delta_{\mu}$ of 13.5, 12.3, 0.04 and 0.25, respectively. Furthermore, the loss factors ($\tan\delta_{\varepsilon}/\varepsilon'$ and $\tan\delta_{\mu}/\mu'$) were calculated to be 0.003 and 0.02 at 0.65 GHz, respectively. These properties make these composite ferrites a choice material for microwave antenna substrate materials in the ultra-high frequency range.

5.4 Reference


CHAPTER 6

SELF-BIASED Y-JUNCTION CIRCULATOR
WITH FERRITES

6.1 Introduction to ferrite based circulator

Barium hexaferrites have been widely used in microwave and millimeter wave devices as permanent magnets and as gyromagnetic materials, e.g., in circulators, filters, isolators, inductors, and phase shifters [1-2]. As a critical component in radar and modern wireless communication systems, it is the microwave circulator that has drawn much attention. Many efforts have been made to design light and miniature circulators with self-biased ferrite materials [3]. We have successfully demonstrated self-biased circulators that operate at Ku-band based on M-type hexaferrites [4]. However, demonstrating self-biased circulators at lower operating frequencies, i.e. X-band, is more difficult due to the need for lower magnetic anisotropy fields and correspondingly lower FMR frequencies. Both W-type and M-type barium ferrite have high saturation magnetization, high Curie temperature, and c-axis anisotropy. However, Ni and Zn substituted W-type barium ferrites may have magnetic anisotropy fields of ~12 kOe and FMR frequencies much lower than those of the M-type ferrites. Furthermore, the Ni-Co or Zn-Co substituted W-type barium ferrites can have anisotropy fields from 4 kOe to 12 kOe with c-axis texture [5-12].
The permeability tensor of the self-biased disk shape hexaferrite materials with large c-axis anisotropy obey the following rules [13-14]:

\[
\begin{bmatrix}
\mu & -jk & 0 \\
jk & \mu & 0 \\
0 & 0 & \mu_0
\end{bmatrix}
\]  \hspace{1cm} (6 - 1)

\[
\mu = \mu_0 \left(1 + \frac{\omega_0 \omega_m}{\omega_0^2 - \omega^2}\right)
\]  \hspace{1cm} (6 - 2)

\[
\kappa = \mu_0 \frac{\omega \omega_m}{\omega_0^2 - \omega^2}
\]  \hspace{1cm} (6 - 3)

\[
\omega_0 = \mu_0 \gamma (H_{app} + H_{ani} - H_{demag})
\]  \hspace{1cm} (6 - 4)

\[
\omega_m = \mu_0 \gamma M
\]  \hspace{1cm} (6 - 5)

where \(M\) is the magnetization, \(\omega\) is the operating frequency, \(\gamma\) is the gyromagnetic ratio, \(H_{app}\) is the applied external magnetic field, \(H_{ani}\) is the uniaxial magnetic anisotropy field, and \(H_{demag}\) is the demagnetization field which affected by the shape of the ferrite material.

In this case, self-biased means no applied external field, and the disk shape makes the value of demagnetization field \((H_{demag})\) equal to that of the remnant magnetization \((M_r)\) in Eqns. (6-4). Also the value of magnetization \((M)\) in Eqns. (6-5) equals to that of the remnant magnetization \((M_r)\) due to the reason of no applied external field. Thus Eqns. (6-4) and Eqns. (6-5) reduced to the following ones:

\[
\omega_0 = \mu_0 \gamma (H_{ani} - M_r)
\]  \hspace{1cm} (6 - 6)

\[
\omega_m = \mu_0 \gamma M_r
\]  \hspace{1cm} (6 - 7)
6.2 Optimization of circulation performance using FEM simulation

The microstrip Y-junction circulator design is adopted in this work. A Duroid® dielectric sheet is used as substrate, which is cut into hexagonal shape with a circular hole in the center of it, the ferrite disk is fit in the circular hole of the substrate. The hybrid substrate is consisted of magnetic material in the center and dielectric material surrounded. The microstrip copper circuit is on the top of this hybrid substrate. The model used for FEM simulation in HFSS® is shown in Fig. 6-1.

![Microstrip Y-junction circulator model with hybrid substrate in HFSS® for simulation.](image)

**Figure 6-1:** Microstrip Y-junction circulator model with hybrid substrate in HFSS® for simulation.

The microstrip Y-junction circulator is a non-reciprocal device used in wireless communications systems. The nonreciprocal property of ferrite materials makes it possible for the transmission and reception of wireless signals occurring at the same time and frequency [4, 15-17]. The circulator in its classic microstrip configuration is a non-
reciprocal 3-port device made from a symmetrically-fed parallel-plate ferrite disk resonator, embedded in an impedance-matching network, so that the propagating wave traveling from port to port encounters low insertion loss in one cyclical direction and high isolation in the other [13].

As one of the critical properties of the circulation performance, low insertion loss is investigated for the effect of magnetic materials in this work. Insertion loss is affected by several magnetic material parameters, such as, saturation magnetization, magnetocrystalline anisotropy field, and ferromagnetic resonance linewidth. Fig. 6-2 shows the simulated results of insertion loss affected by magnetic linewidth, a linear increase of the insertion loss with increase of magnetic intrinsic loss is seen in Fig. 6-2.

![Figure 6-2: Insertion loss with various linewidth (saturation magnetization and anisotropy field are fixed).](image-url)
The simulated results of insertion loss affected by magnetocrystalline anisotropy field is given in Fig. 6-3. The insertion loss decreases with the increase of the magnetic anisotropy field, here the operating frequency is fixed by modifying the dimension of the junction resonator and ferrite disk. It is revealed that in order to achieve low insertion loss, the operating frequency is chosen to be far away from the ferromagnetic resonance frequency, since for a real magnetic sample, the magnetic anisotropy field is fixed as intrinsic property. Comparing Fig. 6-2 and Fig. 6-3, it is shown that magnetic linewidth play a bigger role than magnetic anisotropy field on insertion loss. However during the preparation of self-biased ferrite materials, the tuning of magnetic anisotropy field is much easier than reducing the magnetic linewidth.

![Figure 6-3: Insertion loss with various magnetic anisotropy fields (saturation magnetization and linewidth are fixed).](image)
Figure 6-4: Insertion loss with various saturation magnetizations (magnetic anisotropy field and linewidth are fixed).

Fig. 6-4 shows the simulated results of insertion loss with various saturation magnetizations. Here the value range of the saturation magnetization is decided by real magnetic material properties. It is seen that the change of insertion loss is nonlinear with changed saturation magnetization. In this work, the design of the microstrip Y-junction circulator is used for frequency range from around 8 GHz to 20 GHz, which makes the thickness of dielectric substrate in the dimension of millimeter. Also the magnetic ferrite disk has a large ratio of in plane diameter to the thickness, which leads to a large demagnetization field, the value of the demagnetization field equals to that of the saturation magnetization, so not only the magnetization frequency ($\omega_m$) but also the gyromagnetic frequency ($\omega_0$) are related with saturation magnetization for this structure, thus the
saturation magnetization has a nonlinear effect on insertion loss. However, compared to the effect of magnetic anisotropy field and magnetic linewidth, the effect of saturation magnetization on insertion loss is the smallest.

**Figure 6-5:** Bandwidth pattern with various saturation magnetizations and magnetic anisotropy fields (operating frequency at 10 GHz).

The isolation bandwidth is also a very important characteristic property of circulation performance. Saturation magnetization and magnetic anisotropy field will affect the bandwidth simultaneously, which make the things complicated for study. Fig. 6-5 and Fig. 6-6 show the bandwidth with various saturation magnetizations and magnetic anisotropy fields at operating frequency of 10 GHz and 12 GHz, respectively. It is seen that smaller magnetic anisotropy field and larger saturation magnetization contribute to
good bandwidth, however from the perspective of self-biased magnetic material preparation, it is easier to get material with large magnetic anisotropy and small saturation magnetization than that of small magnetic anisotropy and large saturation magnetization. It is better to know the limitation of the material when pursuing the performance during device design.

**Figure 6-6:** Bandwidth pattern with various saturation magnetizations and magnetic anisotropy fields (operating frequency at 12 GHz).

Fig 6-7 shows a series of S parameters of circulator design with various magnetic anisotropy fields (from 4 kOe to 14 kOe), here the saturation magnetization and magnetic linewidth are fixed. S parameters in Fig 6-7 are optimized for a provided magnetic material,
it is seen that for a real magnetic material the operating frequency is roughly determined by magnetic anisotropy field. The optimized operating frequency is shifted to high frequency range with the increase of magnetic anisotropy field. The detailed parameters with Fig 6-7 are listed in Table 6-1.

**Figure 6-7:** A series of S parameters with various magnetic anisotropy fields (saturation magnetization and magnetic linewidth are fixed).

**Table 6-1:** Detailed information with simulated S parameters with various magnetic anisotropy fields.
In order to explore the effect of dielectric loss on insertion loss of the designed circulator, the decrease of insertion loss at loss tangent from 0.009 to 0.0009 at various magnetic linewidth is shown in Fig. 6-8 and Fig. 6-9. Both dielectric loss and magnetic loss contributed to the insertion loss (electromagnetic loss). When the magnetic linewidth is large with a provided material, the contribution from the dielectric loss is small. However, a small peak is seen in both Fig. 6-8 and Fig. 6-9. The simulated results in both Fig. 6-8 and Fig. 6-9 are based on real material parameters. The magnetic anisotropy field in Fig. 6-8 and Fig. 6-9 are 13 kOe and 8 kOe, respectively. The contribution from dielectric
loss decrease with the increase of magnetic loss of the material below a critical point, and then it will increase a little bit with the increase of magnetic loss, after that it will decrease again with the increase of magnetic linewidth. The values of the critical point various with magnetic materials of different magnetic properties.

Figure 6-8: The decrease of insertion loss at loss tangent from 0.009 to 0.0009 at various magnetic linewidth ($H_A = 13$ kOe).
Figure 6-9: The decrease of insertion loss at loss tangent from 0.0009 to 0.009 at various magnetic linewidth ($H_A = 8$ kOe).

6.3 Measured results for self-biased circulator

In this work, the proposed structure of the microstrip Y-junction for simulation and fabrication is shown in Fig. 6-10. A copper coated Duroid® dielectric substrate with $\varepsilon=2.2$ is first etched in ferric chloride solution to remove the covered copper layer, a round hole with the size of ferrite sample in the center of the substrate is machined, then the ferrite disk was inserted into the hole, and the ferrite sample fits well in the machined hole, dielectric paste may be applied as needed. This hybrid structure served as the substrate of the proposed microstrip Y-junction circulator.

The circuit of the microstrip Y-junction circulator was fabricated by lithography, dielectric substrate with single side copper was coated by photoresist, and then exposed
and developed. The treated substrate was then etched in ferric chloride solution, after this the rest of the photoresist was removed with acetone, and the circuit was finally patterned on the dielectric substrate. The patterned substrate sat on the hybrid substrate with the side of copper pattern faced down, as shown in Fig. 6-10. Since the fixture for the measurement used here has a ground, the ground layer on the bottom of the device in Fig. 6-10 was not fabricated.

**Figure 6-10:** The proposed structure of the microstrip Y-junction for simulation and fabrication.

For the real measurement, the fixture play an role of the convertor from coaxial transmission line to microstrip line, the testing pin in Fig. 6-10 is the internal single line connected to the copper microstrip line circuit, since the diameter of the pin is around 1mm, there is an air gap between the patterned microstrip line circuit and the hybrid dielectric substrate, which will affect the performance of the circulation. When conducting the
measurement, a proper pressure was added on the device to make sure the connection between the signal pin and the microstrip line circuits.

Figure 6-11: The measured magnetic hysteresis loops of the proposed magnetic self-biased material.

The static magnetic properties of provided self-biased magnetic ferrite material were measured by vibrating sample magnetometer (VSM), and the measured magnetic hysteresis loops were shown in Fig. 6-11. The out of plane curve is obtained with the external magnetic field perpendicular to the disk sample plane, while the in plane curve is related with the external magnetic field applied within the disk plane. From Fig. 6-11, an obvious c-axis magnetocrystalline anisotropy is observed. Furthermore, the provided magnetic sample shows a large remnant magnetization in out of plane direction. The
estimated value of magnetocrystalline anisotropy field for the provided sample is around 10-12 kOe, which makes it a good candidate for low operating frequency applications, as we know that the operating frequency is majorly determined by magnetocrystalline anisotropy field.

**Table 6-2:** Detailed information with properties of provided magnetic material and dielectric material.

<table>
<thead>
<tr>
<th>Ferrite properties</th>
<th>Dielectric properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4\pi M_r$</td>
<td>1848 Gauss</td>
</tr>
<tr>
<td>Anisotropy</td>
<td>10000 Oe</td>
</tr>
<tr>
<td>Linewidth</td>
<td>800 Oe</td>
</tr>
<tr>
<td>Diameter</td>
<td>4.08 mm</td>
</tr>
<tr>
<td>Thickness</td>
<td>0.381 mm</td>
</tr>
<tr>
<td>Relative Permittivity</td>
<td>10</td>
</tr>
<tr>
<td>Relative permittivity</td>
<td>2.2</td>
</tr>
<tr>
<td>Loss tangent</td>
<td>0.0009</td>
</tr>
<tr>
<td>Thickness</td>
<td>0.381 mm</td>
</tr>
</tbody>
</table>

The detailed information with properties of provided magnetic material and dielectric materials are shown in Table 6-2, the static magnetic parameters are obtained from hysteresis loop measurement, the magnetic linewidth were measured in a microwave cavity for the ferromagnetic resonance curve. The magnetic sample for the hysteresis loop measurement has a thickness of 2mm. Before it is mounted into the device, the magnetic sample was polished to a thickness of 0.381mm. Since the remnant magnetization is
affected by the thickness of the sample, the value of the remnant magnetization obtained from the hysteresis loop is larger than that of the real disk for circulator.

The simulated S parameters of the proposed microstrip line circulator based on the provided magnetic samples are shown in Fig. 6-12. The measured S parameters indicator a good circulation performance of the microstrip Y-junction circulator at lower operating frequency. In Fig. 6-12, port 1 is the input port, while port 2 and port 3 are the output port, here the signal is transmitting to port 2 and isolated by port 3.

![S parameters graph]

**Figure 6-12:** Simulated S parameters of the proposed microstrip line circulator with a provided magnetic ferrite material.
Figure 6-13: lithography mask of the designed circulator circuit generated from AutoCAD file.
The S parameters of the designed microstrip line self-biased circulator were measured by the Agilent E8364A PNA Series Network Analyzer. For the measurement of this 3-port device, a 50 Ω standard load is connected to one of the ports and the other two ports are connected to the network analyzer as input and output. A picture of the measured S parameters is shown in Fig. 6-13. The measured frequency range is from 8 GHz to 18 GHz, each vertical grid represents 1 GHz, and each horizontal grid represents 10 dB. The peak of the isolation curve is marked to 15.65 GHz.

Figure 6-14: Picture of measured S parameters of the proposed microstrip line circulator with a provided magnetic ferrite material.

The results of the simulation and measurement are summarized in Table 6-3, the measured insertion loss is decreased to 3.7 dB compared to the 2.0 dB from the simulation.
As we mentioned before, there is an unavoidable air gap between the circuit and the substrate with this design for measurement, which is probably response for the worse of insertion loss. The measured operating frequency is also shifted towards higher frequency range. The -20 dB isolation bandwidth is calculated from the simulated and measured curves.

Table 6-3: Comparison of simulated and measured results of circulation performance of the designed circulator.

<table>
<thead>
<tr>
<th></th>
<th>Simulation</th>
<th>Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Insertion Loss</td>
<td>-2.0 dB</td>
<td>-3.7 dB</td>
</tr>
<tr>
<td>Operating Frequency</td>
<td>12.4 GHz</td>
<td>15 GHz</td>
</tr>
<tr>
<td>Bandwidth</td>
<td>590 MHz</td>
<td>500 MHz</td>
</tr>
<tr>
<td>Relative Bandwidth (%)</td>
<td>4.75 %</td>
<td>3.33 %</td>
</tr>
</tbody>
</table>

6.4 Reference


CHAPTER 7

ANTENNA DESIGN BASED ON LOW LOSS FERRITE SUBSTRATE

7.1 Introduction to ferrite substrate for antenna

The magneto-dielectrics materials have two major applications, design of EBG structures and antenna miniaturization. Magneto-dielectric materials can miniaturize both the EBG structures and antenna by the same factor however using moderate values of permittivity and permeability \( n = \sqrt{\varepsilon'\mu'} \) compared to the dielectric-only high permittivity material.

The use of \( \varepsilon' \) identical to \( \mu' \) has two benefits: for multilayer EBG structures, it produces better band-gap rejection levels [1]; for antennas, it allows for ease of impedance matching over a much wider bandwidth. An electrically small antenna may be matched to any frequency by a proper selection of geometry, the use of matching circuits, etc., which may sacrifice antenna efficiency. Thus, we may say the use of \( \varepsilon' \) identical to \( \mu' \) gives flexibility of antenna design.

It has been discussed by Hansen and Burke [2] that the zero-order bandwidth of a simple square shape patch antenna with a magneto-dielectric substrate is given by the following equation:
\[
Bandwidth = \frac{96\sqrt{\mu/\varepsilon} \ t/\lambda_0}{\sqrt{2[4 + 17\sqrt{\mu\varepsilon}]}} \quad (7 - 1)
\]

The item $\sqrt{\mu\varepsilon}$ in Eqns. 7-1 is defined as miniaturization factor, thus for a given $\sqrt{\mu\varepsilon}$ (i.e. the production of the value with real permittivity and real permeability is a constant), a large bandwidth can be achieved by increasing the value of $\mu/\varepsilon$. For the conventional dielectric substrate material ($\mu=1$), the bandwidth of antenna has its own limitation from the aspect of material, however the adoption of magneto-dielectrics materials provides the possibility of increasing the bandwidth from material side.

### 7.2 Simulated results of patch antenna with ferrite substrates

![Figure 7-1: A patch antenna with magneto-dielectric substrate in HFSS® for simulation.](image)
In order to study the performance of the prepared low loss hexaferrite composites in previous chapter used as magneto-dielectric substrate for antenna applications, a simple patch antenna is adopted here. The model of patch antenna with magneto-dielectric substrates for FEM simulation in HFSS® is shown in Fig. 7-1. A copper patch with dimension around 6 cm × 4 cm is layered on the substrate, and the thickness of the substrate is fixed to 0.8 cm in this design. The center pin of the coaxial line is connected via a hole to the copper patch, which is used to feed the patch.

One dielectric only substrate and two magneto-dielectric substrates are applied to the patch antenna to examine the transmission properties, here the dielectric substrate has a real permittivity of 12.5, the magneto-dielectric substrate 1 has a real permittivity of 5 and real permeability of 2.5, and the magneto-dielectric substrate 2 has a real permittivity of 2.5 and real permeability of 5. These three different substrates share the same miniaturization factor ($\sqrt{\mu\varepsilon} = 3.5$), while the value of $\mu/\varepsilon$ is different. The simulated results of return loss with dielectric only and magneto-dielectric substrates are shown in Fig. 7-2. The simulated relative bandwidth of patch antenna with dielectric only substrates is calculated to be 1.3%, which is drawn by black solid line in Fig. 7-2. The simulated relative bandwidth of patch antenna with magneto-dielectric substrate 1 increases to 4.8%, which is drawn by green dash line. Furthermore, the simulated relative bandwidth of patch antenna with magneto-dielectric substrate 2 rises to 8.1%, which is drawn by red dash dot line. It is concluded that the bandwidth for the proposed patch antenna is increasing with the value of $\mu/\varepsilon$. For a dielectric only material, the value of $\mu/\varepsilon$ is much less than 1, only if the use of magneto-dielectric material can increase the value of $\mu/\varepsilon$ to be around 1. It is extremely difficult to make the value of $\mu/\varepsilon$ larger than 1. The prepared low loss
hexaferrite composites in previous chapters have a value of $\mu/\varepsilon$ around 1, which makes the use of magneto-dielectric material possible for antenna substrate. Here the magneto-dielectric material parameters used in simulation are obtained from measured results in previous chapters. For the dielectric only material, the loss tangent of permittivity is set to be 0.001, which is comparable with merchant substrate. For the two magneto-dielectric materials, the loss tangent of permittivity is set to be 0.005, and the loss tangent of permeability is set to be 0.01. The simulate patch antenna shown in Fig. 7-2 is working at 800 MHz. It is seen that the bandwidth increase up to 600% with the introduction of magneto-dielectric substrate.

![Figure 7-2](image)

**Figure 7-2:** Return loss of simulated patch antenna with dielectric only and magneto-dielectric substrates in HFSS®.
The simulated results of far-field radiation situation with dielectric only substrate and magneto-dielectric substrates are given in Fig. 7-3, Fig. 7-4 and Fig. 7-5, respectively. In Fig. 7-3, the patch antenna shows a gain of 1.76 dB with dielectric only substrate. In Fig. 7-4 and Fig. 7-5, the gain of the proposed antenna with magneto-dielectric substrates is 1.49 dB and 1.77 dB, respectively. Since the dielectric only substrate possesses the lowest dielectric loss, it show relatively better radiation performance. For the magneto-dielectric substrates, the one with large $\mu/\varepsilon$ value show a better radiation performance, and at the same time also a larger bandwidth.

![dB(GainTotal)]

Figure 7-3: Gain of simulated patch antenna with dielectric only substrates in HFSS®.
**Figure 7-4:** Gain of simulated patch antenna with magneto-dielectric substrate 1 in HFSS®.

**Figure 7-5:** Gain of simulated patch antenna with magneto-dielectric substrate 2 in HFSS®.
7.3 Reference


CHAPTER 8

CONCLUSIONS

We report the magnetic and structural properties of a series of W-type barium hexaferrites of composition $\text{BaZn}_{2-x}\text{Co}_x\text{Fe}_{16}\text{O}_{27}$ where $x=0.15$, 0.20, and 0.25. The anisotropy field of these BaW ferrites decreased with the substitution of divalent Co ions, while, they maintained crystallographic $c$-axis texture. The measured anisotropy field was $\sim 10$ kOe, and a hysteresis loop squareness $M_r/M_s=79\%$ was obtained due to well-controlled grain size within the range of single domain scale. These two properties make the BaW ferrites suitable for applications in microwave devices at lower frequencies, such as self-biased circulators operated at X-band frequencies.

U-type barium hexaferrite thin films were deposited on (0001) sapphire substrates by pulsed laser deposition. Microstructure and magnetic property of the films were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM) and vibrating sample magnetometer (VSM). Ferromagnetic resonance (FMR) measurements were performed at X-band. The results indicate a measured anisotropy field of $\sim 8$ kOe, and the saturation magnetization ($4\pi M_s$) of 3.6 kG. More interestingly, an optimal post-deposition annealing of the films results in a strong $(0, 0, n)$ crystallographic texture and a high squareness ($M_r/M_s=92\%$) out of the film plane. Furthermore, the highly self-biased ferrite films exhibited low FMR linewidth of $\sim 200$ Oe. The U-type hexaferrite films having
low microwave loss, low magnetic anisotropy field, and high squareness may become an alternative to those Sc or In doped BaM ferrites that were used for self-biased microwave devices at X-band frequencies.

Ferrite composites of the nominal composition $\text{Ba}_3\text{Co}_{2+x}\text{Ir}_x\text{Fe}_{24-2x}\text{O}_{41}$ were studied in order to achieve low magnetic and dielectric losses and equivalent permittivity and permeability over a frequency range of 0.3-1 GHz. Crystallographic structure was characterized by X-ray diffraction, which revealed a Z-type phase accompanied by increasing amounts of Y-type phase as the iridium amount was increased. The measured microwave dielectric and magnetic properties showed that the loss $\tan\delta_\varepsilon$ and loss $\tan\delta_\mu$ were decreased by 80% and 90% at 0.8 GHz with the addition of iridium having $x = 0.12$ and 0.15, respectively. An effective medium approximation was adopted to analyze the composite ferrites having mixed phase structures. Moreover, adding $\text{Bi}_2\text{O}_3$ resulted in equivalent values of real permittivity and real permeability over the studied frequency range. The resultant data gives rise to low loss factors (i.e., $\tan\delta_\varepsilon/\varepsilon' = 0.008$ and $\tan\delta_\mu/\mu' = 0.037$ at 0.8 GHz) while characteristic impedance was the same as that of free space impedance.

A series of Z-type and Y-type ferrite composites with various phase fractions were studied for their RF properties including the measurement of permittivity to permeability spectra over a frequency range of 0.1-10 GHz. Phase identification of the ferrite composites’ constituents was determined by X-ray diffraction. An effective medium approximation was used to predict the magnetic and dielectric behavior of the composites. The experiments indicated that the composite having 75 vol% of Z-type ferrite demonstrated a permeability of ~12 with a near equivalent permittivity, yielding a ratio
(μ’/ε’) of 0.91 at a frequency range from 0.55 to 0.75 GHz. The dielectric loss (i.e., tanδε) and magnetic loss (i.e., tanδμ) were measured to be lower than 0.08 at f =0.1-1 GHz and 0.29 at f =0.1-0.7 GHz, respectively. Furthermore, the loss factors, as tanδε/ε’ and tanδμ/μ’, were calculated to be 0.003 and 0.02 at 0.65 GHz, respectively.