

Handoko Subawi^a* (0), Syoni Soepriyanto^b, Akhmad Ardian Korda^b, Bambang Prijamboedi^c,

Dadan Suhendar^b

 ^aInstitut Teknologi Bandung, Faculty of Mining and Petroleum Engineering, Doctoral Program of Mining Engineering, Jl. Ganesha no. 10, Bandung 40132, Indonesia.
 ^bInstitut Teknologi Bandung, Faculty of Mining and Petroleum Engineering, Department of Metallurgical Engineering, Jl. Ganesha no. 10, Bandung 40132, Indonesia.
 ^cInstitut Teknologi Bandung, Faculty of Mathematics and Natural Sciences, Department of Chemistry, Jl. Ganesha no. 10, Bandung 40132, Indonesia.

Received: November 16, 2021; Revised: March 17, 2022; Accepted: April 10, 2022

In this study, the influence of gadolinium dopant composition on the microwave response performance of yttrium iron garnet was investigated. A hydraulic pressing was used to create the ferrite garnet sample. As the test material, pure oxide powder with an average grain size of 1 micron was employed. A pressure force of 700 MPa was used to compact the sample pellets. The sintering process was carried out in a high-temperature tube furnace at 1450 °C for five hours. This research shows that the calculated findings match the experimental data, demonstrating that the addition of gadolinium reduces the total magnetic moment and insertion loss of yttrium iron garnet. Adding 14 to 25% mol of gadolinium to one mol of iron oxide resulted in an insertion loss value near to zero, according to the calculations. To validate this calculation value, the microwave response of a sample of yttrium iron garnet with 15% mol of gadolinium doping was measured at frequency 4 GHz, yielding an insertion loss value of 0.25 dB, which has a crucial function in lowering the insertion loss value of yttrium iron garnet.

Keywords: Dopant, Gadolinium, Insertion loss, Microwave response, Sintering.

1. Introduction

Ferrite garnets have an important role as a circulator component. Yttrium iron garnet (YIG) has a nearly symmetrical cubic structure, low magnetic loss, and small dielectric loss and is therefore chosen for many applications. The circulator is a device that allows signals entering a port to travel to nearby ports but not to others. The circulator shields the signal source from undesired circuit reflections. Bosma was the first to propose the mechanism of electromagnetic circulation in the circulator¹. Fay and Comstock developed a new approach to circulation that enables the development of narrow band circulators². A circulator design method was presented by Wu and Rosenbaum3. Circulators can be used as switches, multiplexers, dispersion compensators, sensors, etc. The rapid evolution from 4G to 5G telecommunications technology allows new devices to be developed with fast signal response performance. Due to the rapid progress in the development of communication systems, ferrite garnets have become attractive for use in microwave radar and telecommunications equipment.

In the microwave response test, YIG garnets performed well as a high frequency circulator. Microwave response testing was done on YIG compounds with a 9 dB insertion loss value⁴. The sputtering method has been proposed as a revolutionary way of producing YIG, increasing the anticipation that applications would be made in less time and at lower costs⁵. However, of course, the performance of YIG must be improved. The circulator design criterion requires an insertion loss of ~0.8 dB for the 5 GHz frequency. Circulators must be practical, have a minimal insertion loss, and be well-insulated. Physical size downsizing and the utilization of higher operating frequencies are common outcomes of research progress⁶. To construct ferrite devices, microwave circuit theory and magnetic compounds are required7. Microwave device designs and uses in the future will need to be more powerful and lighter. The frequency tuning of this component is obtained from the external magnetic field by a permanent magnet, or by passing a current through the coil. When a higher operating frequency is required, the need for permanent magnets is greater.

The sintering conditions must be understood to reveal the phenomenon of dominant phase development. High heating temperatures (1450+10 °C) are required for yttrium iron garnet (YIG) sintering. When the sintering process is carried out at temperatures below the maximum (1450 °C), the dominant phase of yttrium iron garnet (YIG) is difficult to achieve⁸. High purity iron oxide powder is the main raw material in the fabrication of YIG through the sintering process. Microstructure features such as grain boundaries, secondary

^{*}e-mail: subawihandoko@gmail.com

phases, and intergranular pores contribute significantly to the magnetic properties, which affect the electrical properties and microwave response. An increase in electrical resistivity and a decrease in insertion loss are believed to be correlated as an indicator of the dominant phase.

Phase conversion modeling can be used to confirm the YIG sintering reaction steps, determining the effective pattern of dominant phase formation. The reaction for the creation of YIG garnet is regulated by temperature and time parameters⁹. When the reaction temperature is still low (1300 °C or less), the presence of a perovskite phase will be observed, but with increasing temperature it tends to form a dominant garnet phase. In most of the samples of the YIG formation, yttrium iron perovskite (YIP) and iron (III) oxide (Fe₂O₃) phases were detected. The reaction between Fe₂O₃ and Y₂O₃ has created a YIP layer and is assumed to increase with reaction time and temperature. However, this YIP inhibits heat transfer for further reactions with the Fe₂O₃ particle core, making it difficult to form a YIG dominant phase at relatively low heating temperatures (1300 °C or less).

Paramagnetism is the intrinsic property of certain materials to become temporarily magnetized when placed in an external magnetic field. It is known that gadolinium, iron, cobalt, and nickel are the four elements that can only be magnetized at room temperature. The electron structure of a neutral gadolinium atom is indicated by seven unpaired electrons in the 4f sub shell, which describes the element's strong paramagnetic properties. In the ionized state, the gadolinium ion donates $6s^2$ and $5d^1$ electrons for bonding, leaving the 4f' electron shell intact.

A study of the magnetic properties of certain rare earth oxide doping materials was published where YIG was doped with different rare earth metal cations (Gd, Yb, Ho, and Pr)¹⁰. The sol-gel method was used to prepare the samples. The ferrite garnet peaks were confirmed by XRD analysis, which revealed a cubic phase structure in all samples. At higher frequencies, Pr-doped YIG has improved dielectric and magnetic characteristics, as well as fewer insertion losses because of the ionic radius of Pr³⁺ is smaller than that of Y^{3+} , results in larger grain size increases. In comparison to other YIG and YIG-doped nanoferrites, Gddoped YIG and Ho-doped YIG have reduced the grain sizes. Furthermore, the magnetic studies of doping materials were also investigated11. According to the findings, the saturation magnetization (Ms) parameter of YIG fell with La doping but rose with Nd and Sm doping. For YIG, La-YIG, Nd-YIG, and Sm-YIG, magnetic remanence (Mr) and coercivity (Hc) increased slightly from 37 Oe to 47 Oe, respectively. The findings of the above investigation are consistent with the findings of the magnetic moment parameter analysis¹². At room temperature, gadolinium doping in YIG reduces the magnetic moment per atom.

In this work, efforts to improve the microwave response performance are focused on reducing the values of insertion loss that is measured in decibels (dB). The insertion loss is on the scale of several hundred to a few tenths of a dB and is influenced by the frequency parameter. Although circulators are made to operate over a wide frequency range, they are only made for testing at certain frequencies. Test procedures to obtain accurate measurements of low insertion loss require precise accuracy and are time consuming. In this paper, the results of the calculation of low insertion loss on the composition of gadolinium-doped yttrium iron garnet will be discussed, as evidenced by the results of the microwave response test on several specimen samples.

2. Materials and Methods

The composition of the forming material of a YIG compound with gadolinium doping was studied to analyze the effectiveness of the microwave response. Insertion loss is a key parameter in microwave responsiveness that is being investigated. The microwave response test results from numerous samples of gadolinium doped YIG will be used to validate the value of insertion loss derived from the calculation results. Solid oxides with a purity of > 99.99% were utilized to make the samples, including Gd₂O₂ and Fe₂O₂ (Aldrich[®]) and Y₂O₃ (Supelco®). The addition of gadolinium doping was carefully carried out at a composition of 0%, 15%, and 33% mol against the iron oxide content. The number of molar combinations of Gd₂O₃ and Y₂O₃ compositions was kept constant at 60% mol, while the composition of Fe₂O₂ was kept constant at 100% mol. The concentration of the gadolinium doping substance varies from 0 to 60% mol. The gadolinium content of zero in the pure YIG formula indicates that the YIG compound has not been doped with gadolinium. While gadolinium's 60% mol against iron oxide indicates a pure gadolinium iron garnet combination with all yttrium components replaced by gadolinium.

As illustrated in Figure 1, the solid oxide was weighed and filtered to a 1 μ m size before being put into the available



Figure 1. Ferrite circulator when testing microwave response. (a) Before connection; (b) After connection.

steel dies (K-105[®]) with a 12.5 mm diameter for microwave response testing¹³. The ring-shaped sample has a diameter of 25 mm and is ready for magnetic characterization testing. The sample was introduced into the furnace (F46240CM Thermolyne[®]) and heated to 1450 °C for five hours after being hydraulically pressed. The physical state of the samples was examined visually to determine if they were intact or damaged. The SEM-EDS apparatus (JSM6510A[®]) was used to investigate the crystal morphology. The magnetometer (Remacomp[®]) was used to measure the magnetic characteristics of ferrite materials. Meanwhile, the microwave response, particularly the insertion loss parameter, is measured using the vector network analyzer (Anritzu[®]).

The principles of measuring the insertion loss parameter of a circulator system in this experiment are shown in Figure 2. The S-parameter describes the circulator's reflection in this scenario. The forward transmission coefficient (S12) can be measured with the RF vector network analyzer (Anritzu®). While the notation S12 denotes forward transmission losses, which may be denoted by S12 as P2/P1. The power leakage in the reverse direction, denoted by S13 as P3/P1, is represented by the notation S13. The input power on port 1 [in Watt] is denoted by "P1," whereas the output power on port 2 [in Watt] is denoted by "P2," and the return power is denoted by "P3". The supplied insertion loss (IL) value data sheet can be used to calculate the actual amount of transmitted power loss. It is also possible to determine the behavior of the signal transmission line for various incidence power levels. The insertion loss (IL) [in dB] is defined as:

$$IL = 10 \log_{10} \left(\frac{P_{out}}{P_{in}} \right)$$
(1)

At a frequency of 2 to 5 GHz, insertion loss was measured using a 12.5 mm diameter sample incorporated into the Y-junction circulator casing. The insertion loss test measurement value will be utilized to validate the theoretical calculations' results. The magnetic moment of the YIG compound in the presence of a gadolinium compound at a specific composition is used to calculate the insertion loss value. The value of the test results of the insertion loss will be used to validate the theoretical calculation results. The calculation of the insertion loss value involves the magnetic moment of the YIG compound and the presence of a gadolinium compound at a certain composition. The value of this total magnetic moment has an impact on the acquisition of the insertion loss value. A process flow diagram to evaluate the gadolinium content in yttrium iron garnet is illustrated in Figure 3. To assure the correctness of the calculation findings, the calculation iteration was carried out with a convergence of less than 1%.

3. Results and Discussion

This paper is divided into the following sections to describe a dominant phase Gd-doped YIG compound with low insertion loss. Because of the solid-solid reaction that occurs during the high temperature sintering process, Section 3.1 addresses the morphology of the dominant phase Gd-doped YIG crystals. The goal of this section is to determine the crystal structure of the dominant phase. The approach method for determining the insertion loss value of garnet ferrite as a circulator component is discussed in Section 3.2. The goal of this part is to figure out how gadolinium composition and testing frequency affect the results. The addition of gadolinium doping to garnet ferrite is discussed in Section 3.3. The microwave response characteristic of garnet ferrite is discussed in Section 3.4. The goal of this part is to evaluate the relationship between magnetic characteristics and microwave response performance as a result of variations in gadolinium doping composition.

3.1. Gd-doped YIG crystal morphology

Iron oxide is one of the reactants for the formation of YIG compounds. Because the reactants have not yet experienced a sintering reaction, the effect of one hour of sintering time reveals the presence of the Fe_2O_3 phase. The first 60 minutes of the production process of a yttrium iron perovskite (YIP) as an intermediate phase indicate the dominant value, after which YIP drops substantially. At a temperature of 1420 °C, the effect of the sintering time parameter on the appearance of the YIP phase was noticed¹⁴.

The X-ray diffraction pattern of the YIG sintering experiment, which took place at a temperature of 1450 °C



Figure 2. Diagram of insertion loss measurement principle¹³.



Figure 3. A process flow diagram to evaluate the gadolinium content in yttrium iron garnet.

for five hours, is shown in Figure 4. If the sintering is carried out for less than five hours, a secondary phase will be seen in the form of yttrium iron perovskite. YIP is considered an intermediate product of the solid oxide reaction. YIP will further react with iron oxide to form YIG. The addition of gadolinium is expected to improve the magnetic characteristics and reduce the YIG insertion loss value. Gd-doped YIG exhibits a fine crystalline structure.

The X-ray diffraction equipment used is the Smartlab Rigaku, with the diffraction results printed on paper, from a source of Cu/K α radiation and with a nickel filter. The X-ray diffraction data from the samples was then compared with an ICDD (International Center for Diffraction Data) card. From the X-ray diffraction values that produce the diffraction intensity and angle, analyzed using PANalytical X'Pert HighScore Plus 3.0 to examine the d-spacing parameter of the crystal structure. Figure 4a shows an X-ray diffractogram taken at 2 θ angle (5-90°) to validate the oxides presence in the sample (Fe₂O₃). The diffraction pattern of the YIG test sample identified that the dominant phase in the sample was the crystal structure of yttrium iron garnet at an angle



Figure 4. Diffractogram of a sintered ferrite garnet at 1450 °C, 5 hours.

(2 θ) of 32.3°, 35.5°, and 55.9° with ICDD no. 01-075-0542. Meanwhile, Figure 4b is a diffractogram of gadolinium-doped yttrium iron garnet at 2 θ angle (5-90°). From the results of the X-ray diffraction test, it appears that the dominant phase in the sample is the crystal structure of gadolinium-doped yttrium iron garnet at angles (2 θ) of 32.1°, 35.2°, and 55.6°.

The diffraction pattern of yttrium iron garnet is in accordance with the database diffraction pattern in ICDD no. 01-075-0542. The diffraction pattern (2θ angle, 5-90°) of the sample contained three main diffraction peaks, which corresponded to the diffraction pattern of yttrium iron garnet $(Y_3Fe_5O_{12})$. At an angle of 2 θ which is 32.3°, the diffraction pattern of yttrium iron garnet shows a d-spacing of 2.77396Å. Meanwhile, the diffraction pattern of Gd_{0.75}Y_{2.25}Fe₅O₁₂ indicated the main diffraction peaks according to the diffraction pattern of yttrium iron garnet and Gd_2O_3 . At an angle of 2θ , which is 32.1°, the gadolinium-doped yttrium iron garnet diffraction pattern shows a d-spacing of 2.76887Å. This is consistent with the literature data that the XRD peak shifted gradually to a slightly lower angle (2θ) as the Gd₂O₂ content increased with ICDD no. 98-002-799615. This means that Gd3+ ions occupy the Y³⁺ site, since the ionic radius of Gd³⁺ (0.938Å) is larger than that of Y^{3+} (0.892Å).

The dopant composition added to YIG has an impact on the yttrium oxide composition. The Gd_2O_3 composition added to YIG is always in balance with the Y_2O_3 composition at a total of 60% mol to the Fe_2O_3 composition. The addition of the Gd_2O_3 dopant compound was offset by a decrease in the molarity of the Y_2O_3 composition in YIG. Separate early investigations found that the sintering procedure can only produce a dominant phase if it is carried out for five hours. Table 1 shows the composition of oxide compounds found by observations. The little variation between the estimated and observed values could be attributable to contaminants or the accuracy of the point of observation spot location.

The results of the SEM image of the sample as a result of sintering at 1450 °C showed a compact microstructure. Figure 5a shows a clearly uniform YIG phase as a result of sintering at 1450 °C. Meanwhile, the Gd-doped YIG phase showed denser aggregates. Based on the SEM image obtained, it shows the morphological state of the sample. The effective ionic radius of gadolinium is 0.938 Å, while the ionic radius of iron is 0.55 Å which is smaller. The SEM image shows YIG powder aggregate, but does not indicate a crack. Yttrium and iron compounds form relatively uniform crystals. The voids in YIG are more visible than in Gd-doped YIG, but they appear smaller (red color). Almost all parts of the surface show regular morphology. Another phase (secondary phase) that was different was not observed, meaning that the reaction formed the dominant YIG phase. The smaller sized granules in YIG are seen more than in Gd-doped YIG (yellow color).

Figure 5b shows that Gd-doped YIG aggregates in the presence of gadolinium tend to form crystals with larger but more irregular sizes. The magnitude of the effective ion radius of gadolinium is 0.938 Å as a substitute for yttrium has an effective ionic radius that is almost equal to or slightly larger than the yttrium ion radius of 0.900 Å. This makes the morphology of Gd-doped YIG less void than YIG, but it looks bigger (red color). The smaller particle size in Gddoped YIG looks more reduced with a smaller size compared to YIG (yellow color). The presence of gadolinium in YIG affects the formation of crystal grain growth that looks larger and denser, with a relatively smaller number of voids. Due to sintering, it will increase sample densification, which confirms the massive structure shown by the SEM picture.

The formation of a YIG product occurs through the formation of an intermediate product, namely YIP. Many pores are formed at the grain boundaries at the beginning of sintering. Furthermore, there is a decrease in the number of pores formed so that the density value increases. A longer sintering process will reduce the number of pores formed. Some of the raw ingredients (Fe₂O₃) have not fully reacted when the sintering process begins in the first hour. An XRD diffractogram can be used to identify the iron oxide component. The following pattern can be used to represent the reaction mechanism that occurs:

Table 1. The oxide composition in yttrium iron garnets.

Compound(s)	Solid oxide	calculated (% w)	spot_1 (% w)	spot_2 (% w)	spot_3 (% w)	spot_4 (% w)
Gd _{0,75} Y _{2,25} Fe ₅ O ₁₂	Gd_2O_3	16.1	16.7	16.5	16.1	15.3
	Y_2O_3	33.0	39.2	40.7	40.4	40.6
	Fe ₂ O ₃	50.8	44.1	42.8	43.5	44.2
Y ₃ Fe ₅ O ₁₂	Y ₂ O ₃	45.9	47.1	48.8	48.1	47.9
	Fe ₂ O ₃	54.1	52.9	51.2	51.9	52.1



Figure 5. SEM images of Y₃Fe₅O₁₂ and Gd_{0.75}Y_{2.25}Fe₅O₁₂.

$$1.5(1-x) Fe_2O_3 + 1.5(1-x) Y_2O_3 \leftrightarrow 3(1-x) YFeO_3$$
(2)

$$1.5x Fe_2O_3 + 1.5x Gd_2O_3 \leftrightarrow 3x GdFeO_3$$
(3)

$$3(1-x) Fe_2O_3 + 3(1-x) YFeO_3 \leftrightarrow (1-x) Y_3Fe_5O_{12} + 2(1-x) Fe_2O_3 \qquad (4)$$

$$3x Fe_2O_3 + 3x GdFeO_3 \leftrightarrow x Gd_3Fe_5O_{12} + 2x Fe_2O_3$$
(5)

$$x \ Gd_3Fe_5O_{12} + (1-x) \ Y_3Fe_5O_{12} \to Gd_{3x}Y_{3(1-x)}Fe_5O_{12}$$
 (6)

$$4.5 \ Fe_2O_3 + \ 1.5(1-x) \ Y_2O_3 + \ 1.5x \ Gd_2O_3 \rightarrow \ Gd_{3x}Y_{3(1-x)}Fe_5O_{12} + \ 2 \ Fe_2O_3 \tag{7}$$

Furthermore, after three hours of sintering, an intermediate compound in the form of yttrium iron perovskite was discovered among the garnet products. From the use of doping materials, gadolinium iron perovskite intermediates will result. The following pattern can be used to explain the reaction mechanism:

$$4(1-x) Fe_2O_3 + 4(1-x) Y_2O_3 \leftrightarrow 8(1-x) YFeO_3$$
(8)

$$4x \ Fe_2O_3 + \ 4x \ Gd_2O_3 \leftrightarrow 8x \ GdFeO_3 \tag{9}$$

$$(1-x) Fe_2O_3 + 8(1-x) YFeO_3 \leftrightarrow (1-x) Y_3Fe_5O_{12} + 5(1-x) YFeO_3$$
 (10)

$$x Fe_2O_3 + 8x GdFeO_3 \leftrightarrow x Gd_3Fe_5O_{12} + 5x GdFeO_3$$
(11)

$$(1-x) Y_3 Fe_5 O_{12} + x Gd_3 Fe_5 O_{12} \rightarrow Gd_{3x} Y_{3(1-x)} Fe_5 O_{12}$$
(12)

$$5 Fe_2O_3 + 4(1-x) Y_2O_3 + 4x Gd_2O_3 \rightarrow Gd_{3x}Y_{3(1-x)}Fe_5O_{12} + 5(1-x) YFeO_3 + 5x GdFeO_3$$
(13)

When the sintering process reaches a minimum of five hours, the sintering reaction condition enters a stable state. The only material created is yttrium iron garnet. The use of doping materials will result in a gadolinium-doped iron garnet as the final product. The following pattern can be used to represent the reaction mechanism that occurs:

$$1.5(1-x) Fe_2O_3 + 1.5(1-x) Y_2O_3 \leftrightarrow 3(1-x) YFeO_3$$
(14)

$$1.5x \ Fe_2O_3 + 1.5x \ Gd_2O_3 \leftrightarrow 3x \ GdFeO_3 \tag{15}$$

$$(1-x) Fe_2O_3 + 3(1-x) YFeO_3 \leftrightarrow (1-x) Y_3Fe_5O_{12}$$
(16)

$$x Fe_2O_3 + 3x GdFeO_3 \leftrightarrow x Gd_3Fe_5O_{12}$$
(17)

$$(1-x) Y_3 Fe_5 O_{12} + x Gd_3 Fe_5 O_{12} \to Gd_{3x} Y_{3(1-x)} Fe_5 O_{12}$$
(18)

$$5 Fe_2O_3 + 1.5(1-x)Y_2O_3 + 1.5x Gd_2O_3 \rightarrow Gd_{3x}Y_{3(1-x)}Fe_5O_{12}$$
(19)

The oxide compounds are rearranged during the sintering process to generate a tighter structure. Temperature and sintering time are mostly responsible for changes in the layout of pores that are getting progressively dense. When the sintering period reaches five hours, the crystal density achieves a stable state, where the crystals can organize themselves into a density of more than 70%. The measurement results of densification parameters on the sample of Gd_{0.75}Y_{2.25}Fe₅O₁₂ showed values of 2.18%, 73.29%, and 75.41% as the result of sintering at 1450 °C for 1 hour, 3 hours, and 5 hours, respectively. Densification is defined as the ratio between the actual density shrinkage of the sample compared to the theoretical density shrinkage. Density measurements were carried out by measuring the weight of the solid oxide against the pellet volume before and after sintering. However, the size of the gadolinium ion influences the compaction process, with a minor drop in density when gadolinium doping is increased to close to 60% mol. According to the testing results, a five-hour sintering time at 1450 °C proved in getting a dominant phase of the ferrite garnet product. This product with the dominant phase is preferred in this study since it has been shown to have the best microwave response. The microwave response performance will be reduced if intermediary components are mixed into the product.

The metal-organic breakdown approach was used to conduct the morphological investigation, which was based on YIG micrographs¹⁶. At a temperature of less than 1300 °C, the tendency for morphological changes in the form of irregularly formed granules was detected. From a state of fluency, individual particles resemble prolate spheroids, and some are connected to one another.

3.2. Insertion loss of Gd-doped YIG

Understanding the magnetic characteristics of ferrite garnets is the first step in understanding microwave response. The presence of gadolinium cations in the dodecahedral position (c) of the $Gd_{3x}Y_{3(1-x)}Fe_5O_{12}$ molecule will substitute for yttrium during the production process. Because of the interaction between iron ions at position (a) and iron ions at position (d), a magnetic moment opposite to the total magnetic moment occurs. Due to the inclusion of a gadolinium ion in the 24c position in place of the yttrium ion, the total magnetic moment of gadolinium-iron-garnet (GdIG) or $Gd_{3}Fe_{5}O_{12}$ is equivalent to 32 Bohr magnetons (μ_{B}), due to the existence of a gadolinium ion in place of the yttrium ion at the 24c position. Iron ions occupy two sublattices that are antiparallel. The magnetic moment of the two positions, 24c and 16a, is (2 Fe³⁺)(5 $\mu_{\rm B}$ /Fe³⁺) or 10 $\mu_{\rm B}$, with position 24d, which has more iron ions, determining the direction of the magnetic moment. The net magnetic moment (μ_{net}) of the $Gd_{3x}Y_{3(1-x)}Fe_5O_{12}$ combination can be represented in the following equation:

$$\mu_{net} = \left[\left\{ 3(1-x)nY^{3+} + 3(x) nGd^{3+} \right\} + \left\{ 2 nFe^{3+} \right\} + \left\{ 3 nFe^{3+} \right\} \right] \mu_B \quad (20)$$

where x is the fraction of gadolinium cations that substitute for yttrium cations and n is the number of unpaired electrons in each trivalent ion.

The effect of temperature on the magnetic moments of the gadolinium cations is greater than that of the iron cations. At a certain temperature, the magnetic moments of gadolinium and

iron cations are the same, that is, at a temperature called the compensation temperature. The compensation temperature at which M equals zero, for gadolinium-iron-garnet, is 15 °C. If the number of gadolinium cations decrease while yttrium cations increase, the compensation temperature will change. Next, this will change the magnetization and line width parameters. The Curie temperature occurred at 290 °C for all compositions.

YIG is an important material as a component of radar and telecommunications equipment. The insertion loss parameter can be used to evaluate the material's microwave response performance. The amount of power loss conveyed is equal to the insertion loss (IL) value. This aids in determining the performance of signal transmission lines at various incident power levels. The composition of the constituent compounds can be used to evaluate the microwave response performance of Gd-doped YIG. Gadolinium is the doping chemical used to engineer the performance of YIG in this article.

The evaluation of the total magnetic moment is the first step in studying the Gd-doped YIG microwave response. The insertion loss value can then be computed. The wave signal is reduced by the insertion loss parameter after passing through the YIG material system. This insertion loss parameter can be computed using the Equation 21 for the ratio of charged to uncharged power. A microwave system's power ranges from 1 femtowatt (10-15 watt) to 1 megawatt (10⁶ watts). Electrical power, measured in watts, is the strength or amplitude of electromagnetic waves. Because microwave power ranges so widely, it's easier to compute in dB and dBm units. This is why dB and dBm are used in all microwave measurements. The dB notation will reduce the broad range of microwave power values found in microwave components to a more manageable level. It is feasible to combine picowatts and megawatts in a single calculation using this dB and dBm approach. The conversion of decimal integers to the dB unit is shown in Table 2. The decimal system is in column one, scientific notation is in column two, and the dB system is in column three.

Laboratory tests or a computational approach can be used to determine the performance of the YIG microwave response when dopant chemicals are added. Insertion loss is the microwave response parameter that will be employed in the evaluation. The optimum composition of dopant compounds in ferrite garnet can be determined using this calculating approach method. The ferrite garnet in question

Table 2. The conversion of decimal numbers to dB unit.

Decimal numbers	Scientific notation	dB		
0.0001	10-4	-40		
0.001	10-3	-30		
0.01	10-2	-20		
0.1	10-1	-10		
1	10^{0}	0		
10	10 ¹	10		
20	101.3	13		
50	101.7	17		
100	10 ²	20		
1000	10 ³	30		
10000	10^{4}	40		

is a YIG doped with gadolinium. The following insertion loss parameters (in decibels) can be calculated using the following mathematical equation¹⁷:

$$IL = 10 \log \left(\frac{Q_L}{1 - \frac{Q_L}{Q_U}} \right)$$
(21)

where IL is the insertion loss, Q_L is the loaded power, and Q_{U} is the unloaded power.

The Q_L parameter is obtained through the following equation:

$$Q_{L} = \left(\frac{\begin{pmatrix} k & r_{eff} \end{pmatrix}^{2} - 1}{2\sqrt{3}(K/\mu)}\right)$$
(22)

where: k is the number of wave propagation, r_{eff} is the effective radius of the ferrite disk, K is the off-diagonal component of μ , and μ is the diagonal component of the permeability tensor.

Next, the parameter of $1/Q_U$ is obtained by rearranging the equation according to the following:

$$\frac{1}{Q_{\rm U}} = \left(\frac{\gamma^2 \times \left(4\pi \ M_{\rm s}\right) \times \Delta H}{2 \times \omega^2} + \tan \ \delta\right)$$
(23)

where γ is the magnetization frequency, M_s is the magnetization, ΔH is the ferrite line width (Oersted), ω is the driving frequency of the radio frequency (radians, $\omega = 2\pi f$), tan δ is the dielectric loss tangent.

The magnetic moment parameter is used to obtain the saturation magnetization. Calculation of saturation magnetization is obtained through the following equation:

$$Ms = \frac{\underset{net}{\overset{\mu}{\underset{cell unit volume}{}}} \times Bohr magneton}{cell unit volume}$$
(24)

where μ_{net} will be -5.00 when the gadolinium composition is equal to 0% mol, while μ_{net} will be 15.75 when the gadolinium composition is 60% mol.

The yttrium iron garnet combination is composed of 60% mol yttrium oxide and 1 mol iron oxide. The presence of gadolinium will cause yttrium to change its location. The composition of the yttrium oxide and gadolinium oxide compounds is kept consistent to maintain the soft ferrite features of yttrium iron garnet. Figure 6 below shows the magnetic moments of yttrium and gadolinium ions in the Gd-doped YIG compound.

Frequency (GHz)	Insertion loss of YIG tested (dB)	Insertion loss of YIG calc. (dB)	Convergence	Criteria	Ref
4	5.51	5.51	0.08%	< 1.00%	this work
10	5.00	4.98	0.38%	< 1.00%	Wu ¹⁰
20	3.90	3.88	0.40%	< 1.00%	He et al. ¹¹
28	2.80	2.79	0.49%	< 1.00%	Harris ¹²

Table 3. The tested and calculated value of YIG insertion loss.



Figure 6. Magnetic moments of yttrium and gadolinium ions in the Gd-doped YIG compound.

Furthermore, to ease the calculation of insertion loss, the above equation needs to be simplified into the following equation:

$$\frac{1}{Q_{\rm U}} = \frac{M_{\rm s}}{C_{\rm IL}} \tag{25}$$

where Q_U is the unloaded power, M_s is the magnetization, and C_{IL} is the calculation initiation value.

The calculation will be reduced even more by using the approach where *K* is equal to 0.364 and μ is equal to 0.967. The effective radius definition is utilized to rectify the previously known ferrite radius parameter. The simplification assumes that the tan δ effect is negligible. For frequencies between 1 and 30 GHz, an accuracy threshold of less than 1% is used in the calculation. The calculation was done to see how gadolinium doping affects the microwave response of yttrium iron garnet (YIG) compounds. Table 3 shows the measurement findings as well as the YIG insertion loss value computation.

3.3. Gd-doped YIG formulation

The composition of the doping material influences the value of the magnetic moment, which is then utilized to compute the insertion loss value of the YIG compound. As a reference for YIG properties, the initial condition of a pure YIG compound without doping was employed. When gadolinium is doped, a non-stoichiometric molecule called Gd-doped YIG is formed. Gadolinium's maximal doping limit is 60% mol to 1 mol of iron oxide, resulting

in gadolinium iron garnet (GdIG), in which all the yttrium has been entirely replaced by gadolinium.

When the magnetization value drops, the insertion loss value falls as well, eventually reaching zero. The total magnetic moment is multiplied by the Bohr magneton number and divided by the volume of the YIG unit cell to get the saturation magnetization value. The unit cell volume is 1.9E-27 m3, and the Bohr magneton value used to calculate saturated magnetization is 9.27E⁻²⁴ A•m². Another important factor is frequency, which affects the value of the magnetic moment. When the frequency is lower, the value of the magnetic moment decreases. The total magnetic moment tends to correspond with an insertion loss of close to zero at a given dopant composition. The changes in the value of insertion loss as a function of magnetic moment and dopant composition are shown in Figure 7. Meanwhile, Table 4 shows the insertion loss parameter calculation value for Gd-doped YIG compounds at varied doping compositions and operating frequencies.

The insertion loss value changes gradually as the applied operating frequency is varied. A statistical data set was created based on data taken from the literature and paired with experimental data to see the trend of the frequency parameter's influence on the insertion loss value achieved. The insertion loss value obtained will drop slightly as the applied frequency increases. The insertion loss value will be reduced across a larger range at higher frequencies. Insertion loss is lowest at a frequency of roughly 4 GHz when gadolinium concentrations are between 15% and 25% mol for 1 mol of iron oxide. The range of dopant compositions required to produce near-zero insertion loss will be broader at higher frequencies.

The insertion loss value derived using this calculation approach will subsequently be validated using data collected from laboratory testing of the microwave response of samples. In fact, achieving a zero number for the insertion loss measurement is almost unattainable. This is conceivable due to the influence of each component that makes up the circulator's integration factor prior to laboratory testing. This integration factor is quite delicate, and it's challenging to obtain optimum conditions where all components can be placed firmly and compactly enough to produce zero insertion loss. The lowest insertion loss value from the laboratory test results in this experiment was 0.24 dB.

3.4. Microwave response performance

The dopant composition in the yttrium iron garnet (YIG) compound has a direct impact on the material's magnetic moment value. If the composition of the dopant compound in YIG changes, the magnetic moment will change. Iron cations occupy two crystallographic positions in the YIG crystal structure, namely tetrahedral and octahedral. The addition of

	Gadolinium composition. in mol/mol Fe ₂ O ₃						
Freq. (GHz)	0%	10%	20%	30%	40%	50%	60%
	Insertion loss of Gd-doped YIG (dB)						
4	5.51	2.47	0.00	2.69	5.62	7.35	8.48
10	4.98	2.16	0.00	1.48	4.64	6.45	7.61
20	3.88	1.56	0.00	0.00	2.13	4.22	5.51
28	2.79	1.04	0.00	0.00	0.00	1.03	2.63

Table 4. The calculation results of the insertion loss value.



Figure 7. Changes of the value of insertion loss to magnetic moment (a), dopant composition (b).

gadolinium (0.935 Å) to the YIG molecule at the dodecahedral position will occupy the yttrium lattice (0.900 Å). In other circumstances, comparable atoms such as silicon (0.40 Å), germanium (0.53 Å), nickel (0.69 Å), cobalt (0.74 Å), and manganese (0.67 Å) can replace ferrous cations (0.55 Å) in YIG at the tetrahedral location. Magnesium (0.72 Å), indium (0.80 Å), and Scandinavian (0.74 Å) on the other hand, have slightly greater radii than iron and thus occupy octahedral locations, which might increase the total magnetic moment.

The sum of the magnetic moments of the number of unpaired electrons of the yttrium, gadolinium, and iron ions yields the total magnetic moment of the Gd-doped YIG. An electron has a magnetic moment of about one Bohr magneton. The Bohr magneton is a physical constant and a unit of measure for an electron's magnetic moment as a result of its orbital angular momentum, or spin. The total magnetic moment and saturation magnetization are proportional to changes in the value of insertion loss. A reduction in the saturation magnetization value causes the insertion loss value to drop. The addition of rare earth metal can impact the decrease in saturation magnetization value. The inclusion of rare earth metal dopants into yttrium at the dodecahedral position, which reduces the total magnetization of ferrous ions, can influence the decrease in saturation magnetization value. In other words, doping gadolinium into YIG to generate the $Gd_{3x}Y_{3(1-x)}Fe_5O_{12}$ molecule can lower the saturation magnetization value. The gadolinium content in $Gd_{3x}Y_{3(1-x)}$ Fe₅O₁₂ ranges from 0 to 60% mol to 1 mol Fe₂O₃ in this calculation. Figure 8 depicts the effects of dopant content on

coercive force and saturation magnetization. The coercivity value increases slightly as the doping composition increases, but it rapidly declines when the doping content exceeds roughly 15% mol. The saturation magnetization value tends to be at a minimum at 15% mol doping composition. Between calculations and measurements, there is a similar trend that produces a minimum curve in the composition.

According to the calculation method, the addition of a modest amount of dopant compounds will minimize the insertion loss value. The value of a ferrite material's magnetic moment can be traced back to changes in its insertion loss. A somewhat higher insertion loss value will result from a lower testing frequency. The correlation of the value of insertion loss to coercive force, and saturation magnetization at 4 GHz is shown in Figure 9. As shown in this graph, the insertion loss value increases slightly when the coercivity value decreases. Meanwhile, the insertion loss value tends to decrease as the saturation magnetization value drops and achieves a minimum of 15% mol doping composition. Between calculations and measurements, there is a similar trend that produces a minimum curve in the composition. This study resulted in the lowest insertion loss value of 0.25 dB. The insertion loss value found in this study appears to be superior to those reported in the literature¹⁸⁻²⁵. This lower insertion loss value indicates that the ferrite garnet sample obtained has a higher phase purity, meaning that the secondary phase is relatively very low or practically non-existent.

The coercive strength (Hc) of the Gd-doped YIG compound in this investigation was 0.53 Oe, which is significantly better



Figure 8. Changes in dopant composition to coercive force (a) and saturation magnetization (b).



Figure 9. Correlation of the value of insertion loss to coercive force (a) and saturation magnetization (b).

than that reported in prior studies based on magnetic property measurements^{26,27}. In addition, the calculated magnetization value was validated by the results of the YIG sample test with gadolinium doping at compositions of 0%, 14%, and 33% mol. The addition of gadolinium doping decreased the magnetization value at low compositions, but the addition of more than 40% mol of gadolinium tended to raise it. The saturation magnetization will decrease as the measurement temperature rises^{28,29}. The addition of too much gadolinium will emphasize the basic features of gadolinium oxide, which is not categorized as a soft ferrite and can be detected by an increase in the insertion loss parameter. As a result, the gadolinium doping is chosen so that the properties of the resulting compound have the lowest insertion loss value, as this parameter is advantageous in microwave technology applications.

4. Conclusion

The gadolinium ion goes to the yttrium location in the dodecahedral lattice during the sintering reaction of Gd-doped YIG in the solid state. The Gd-doped YIG sintering procedure resulted in a solid sintered product with a dominant phase and no secondary phase after five hours at 1450 °C. The modest composition of YIG with the addition of gadolinium has been proven to reduce insertion loss considerably. On the other hand, too much gadolinium doping will emphasize the fact that gadolinium oxide is not a soft ferrite, raising the insertion loss parameter value. According to the calculations, adding a gadolinium composition in the range of 15% to 25% mol to 1 mol Fe₂O₃ in the YIG will effectively result in a low insertion loss close to zero. When compared to the real value of the laboratory test results, this formula yields the lowest insertion loss value of 0.25 dB. Because of the adjustment factor, obtaining the absolute zero insertion loss number from laboratory test results is challenging in practice. The component integration element of the circulator correction factor is highly delicate.

5. Acknowledgments

This research is part of dissertation research of H Subawi funded by the ITB through PPMI Grant No. 18-IT1.C05-SK-OT.00-2021. The authors also thank Mr. Febiandra Eka, Mr. Danu Anjasmoro, and Mr. Komang Widhi Widantha for their assistance in data collection used in the experiment.

6. References

- Bosma H. On stripline Y-circulation at UHF. IEEE Trans Microw Theory Tech. 1964;12(1):61-72.
- Fay C, Comstock R. Operation of the ferrite junction circulator. IEEE Trans Microw Theory Tech. 1965;13(1):15-27.
- Wu YS, Rosenbaum FJ. Wideband operation of microstrip circulators. IEEE Trans Microw Theory Tech. 1974;22(10):849-56.
- Manuilov SA, Fors R, Khartsev SI, Grishind AM. Pulsed laser deposited Y₃Fe₃O₁₂ films for magnetostatic wave band pass filters. Solid State Phenom. 2009;152-153:377-80.
- Alyami MFA. Improving the structural and magnetic properties of Yttrium Iron Garnet [thesis]. Leeds: University of Leeds; 2021.
- Adam JD, Davis LE, Dionne GF, Schloemann EF, Stitzer SN. Ferrite devices and materials. IEEE Trans Microw Theory Tech. 2002;50(3):721-37.
- Özgür U, Alivov Y, Morkoc H. Microwave ferrites, part 1: fundamental properties. J Mater Sci Mater Electron. 2009;20(9):789-834.
- Ali WFF, Othman MA, Ain MF, Abdullah NS, Ahmad ZA. Studies on the formation of yttrium iron garnet through stoichiometry modification prepared by conventional solid-state method. J Eur Ceram Soc. 2013;33(7):1317-24.
- Praveena K, Srinath S. Effect of Gd³⁺ on dielectric and magnetic properties of Y₃Fe₅O₁₂. J Magn Magn Mater. 2014;349:45-50.
- Akhtar MN, Yousaf M, Khan SN, Nazir MS, Ahmad M, Khan MA. Structural and electromagnetic evaluations of YIG rare earth doped (Gd, Pr, Ho,Yb) nanoferrites for high frequency applications. Ceram Int. 2017;43(18):17032-40.
- Lau ZY, Lee KC, Soleimani H, Beh HG. Experimental study of electromagnetic-assisted rare-earth doped Yttrium Iron Garnet (YIG) nanofluids on wettability and interfacial tension alteration. Energies. 2019;12(20):3806.
- Kuila M, Reddy VR, Srinath S, Phase DM. Study of gadolinium (Gd) doped epitaxial yttrium iron garnet (YIG) thin films. AIP Conf Proc. 2020;2265(1):030529.
- Valvo. Insertion loss measurement. Hamburg; 2017. (Application Note; no. ANV005).
- Zhao C, Li Y, Shen X, Cao Z, Cao Z, Wen Z, et al. Studies on highly dense, pure YIG polycrystalline ceramics fabricated by the tape-casting method. Research Square. 2020. In press. https://doi.org/10.21203/rs.3.rs-57431/v1.
- Patel SKS, Lee JH, Bhoi B, Lim JT, Kim C, Kim SK. Effects of isovalent substitution on structural and magnetic properties

of nanocrystalline $Y_{3-x}Gd_xFe_5O_{12}$ ($0 \le x \le 3$) garnets. J Magn Magn Mater. 2018;452:48-54.

- Liu Y, Wang X, Zhu J, Huang R, Tang D. Structure dependence of magnetic properties in yttrium iron garnet by metal-organic decomposition method. Chin Phys B. 2017;26(5):057501.
- Genner R. The design of microstrip circulators. Malvern: Royal Radar Establishment; 1976. 296 p.
- Wu J. Planar Tunable RF/Microwave devices with magnetic, ferroelectric, and multi-ferric materials [dissertation]. Massachusetts: Northeastern University of Boston; 2012.
- He Y, He P, Dae Yoon S, Parimi PV, Rachford FJ, Harris VG, et al. Tunable negative index metamaterial using yttrium iron garnet. J Magn Magn Mater. 2007;313(1):187-91.
- Harris VG. Modern microwave ferrites. IEEE Trans Magn. 2012;48(3):1075-104.
- Truong NK, Lee DS, Kim SJ, Lee M, Hwang KC, Yang Y, et al. SPDT switch with body-floating technique and an ultra-small active matching network using an on-chip solenoid inductor for BLE applications. Electronics. 2018;7(11):1-11.
- Pintus P, Huang D, Morton PA, Shoji Y, Mizumoto T, Bowers JE. Broadband TE optical isolators and circulators in silicon photonics through Ce-YIG bonding. J Lightwave Technol. 2019;37(5):1463-73.
- Sun M, Han X, Chen S. Synthesis and photocatalytic activity of a nano-cobalt ferrite catalyst for the photo-degradation of various dyes under simulated sunlight irradiation. Mater Sci Semicond Process. 2019;91(1):367-76.
- Tatarenko AS, Bichurin MI. Microwave magnetoelectric devices. Adv Condens Matter Phys. 2012;2012:286562. http://dx.doi. org/10.1155/2012/286562.
- Popov MA, Zavislyak IV, Srinivasan G. A magnetic field tunable yttrium iron garnet millimeter-wave dielectric phase shifter: theory and experiment. Progress in Electromagnetics Research C. 2012;25:145-57.
- Serra RA, Ogasawara T, Ogasawara AS. Detailed crystallization study of co-precipitated Y_{1.47}Gd_{1.53}Fe₅O₁₂ & relevant magnetic properties. Quim Nova. 2007;30(7):1545-9.
- Mallmann EJJ, Sombra ASB, Goes JC, Fechine BAP. Yttrium iron garnet: properties and applications review. Solid State Phenom. 2013;202:65-96.
- Likhart DK. Microwave circulator design. 2nd ed. Norwood: Artech House Inc.; 2014. 364 p.
- Hench LL, West JK. Principles of electronic ceramics. 1st ed. New York: John Wiley & Sons; 1990. 576 p.