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Magnetic and Microwave Properties of Polycrystalline Gadolinium Iron Garnet

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Abstract. The microwave loss in nanosized GdIG particles synthesized using mechanical alloying technique was investigated. There were very few of research on the microwave properties of nanosized particle GdIG and there is no attempt investigating on the material at C-band frequency range (4-8 GHz) and its correlation with the microstructure. Gadolinium (III) oxide and iron (III) oxide, αFe_2O_3 were used as the starting materials. The mixed powder was then milled in a high-energy ball mixer/mill SPEX8000D for 3 hours. The samples were sintered at temperature 1200 ^oC for 10 hours in an ambient air environment. The phase formation of the sintered samples was analyzed using a Philips X'Pert Diffractometer with Cu-Kα radiation. Complex permeability consisted of real permeability and magnetic loss factor were measured using an Agilent HP4291A Impedance Material Analyzer in frequency range from 10 MHz to 1 GHz. A PNA-N5227 Vector Network Analyzer (VNA) was used to obtain the information on ferromagnetic linewidth broadening, ΔH that represents the microwave loss in the samples in frequency range of 4 to 8 GHz (C-band). The ΔH value was calculated from the transmission (S_{21}) data acquired from VNA. The single phase GdIG showed low initial permeability of 1.48 and low magnetic loss of 0.13 when applied with low frequency range energy (10MHz - 1GHz). From these data, it is validated that GdIG is a suitable material for microwave devices for high frequency range.

Introduction

In recent years, a vast number of microwave devices such as circulator, phase shifter, isolator and miniaturized antenna, which requires an extremely low microwave loss had been contrived extensively due to high demand in microwave and magneto-optical industry [1]. In order to achieve extremely low microwave loss requirement, material selection and processing method are aspects to be scrutinized. Among type of ferrites, garnet-type ferrites are well-known low microwave loss materials due to their superior properties such as high resistivity, moderate permeability, and low eddy current loss [2]. Lately, yttrium iron garnet (YIG) ferrite has been extensively studied and chosen as the best candidate to be exploited as a passive microwave component. Gadolinium iron garnet (GdIG) also has remarkable properties that makes it suitable, not only for microwave application, but also in high-density magnetic, magneto-optical information storage and cryogenic magnetic refrigeration applications [3]. Properties of garnet is strongly affected by crystal structure and microstructure. Nanosized particles show a novel properties as compared to bulk counterpart [4]. Therefore, there are many approaches in previous literatures carried out by researchers to obtain nanosized powders that lead to desired properties. The mechanical alloying is a solid-state reaction method that allows one to attain nanosized powder. The capability to produce a large amount of samples and an effective time consuming method makes this method favourable for industrial purposes. Although chemical method such as sol-gel, co-precipitation etc. is a promising method to obtain a homogeneous and nanosized powder, it can only produce small amount of powder which is not applicable in larger scale production. Besides, the likelihood of formation of intermediate phase is rather high. This study aimed to study the microwave loss in nanosized particles GdIG synthesized using the mechanical alloying technique in order to discover on how the microstructure would affect the microwave properties.

Materials and Methods

Preparation of sample. Raw materials of gadolinium(III) oxide, Gd_2O_3 (99.99%), and hematite, α-Fe_2O_3 (99%) both from Alfa Aesar were weighed according to the following stoichiometric formula in Eq. 1:

$$3Gd_2O_3 + 5\alpha\text{-}Fe_2O_3 \longrightarrow 2Gd_3Fe_5O_{12} \quad (1)$$

These powder were mixed and ground in a mortar until the mixture became homogeneous. Then, the mixture was milled in a SPEX8000D mixer/mill with 1425 rpm for 3 hours in steel vials. The process was carried out in air and both vials were closed during the process. The mixture was milled with ball to powder ratio of 10:1. The milled powder was allotted to 3 parts for the characterization purpose; powder form, toroid and pellet. The powder for toroid and pellet was mixed with organic binder of 1wt.% polyvinyl alcohol and pressed under a pressure of 3 tonne. The three samples were then sintered in an Elite box-type furnace at 1200 ^oC with the heating rate of 4 ^oC/min for 10 hours in an ambient air environment. For the microwave test, the toroid sample was cut beforehand into a cubic shape of 2 mm and inserted into an home made equipment called air-driven mill to form a spherical sample with ~2 mm diameter.

Characterization of sample. The phase formation was identified using X'Pert Highscore software from the data collected at room temperature using Philips X'Pert Diffractometer with Cu-Kα radiation source of wavelength, λ=1.54060Å, in the range of 2θ^o-8θ^o. The grains formed after sintering were observed using a Nova-Nano 230 Field Emission Scanning Electron Microscope (FESEM) and 200 grains size were measured with the aid of ImageJ software. Complex permeability data consisted of real and imaginary permeability were acquired from an Agilent HP4291A Impedance Material Analyzer in the frequency range from 10 MHz to 1 GHz. For microwave properties measurement, a PNA-N5227 Vector Network Analyzer was used where a spherical sample was placed in a cylindrical cavity resonator, and measured at microwave frequency of C-band (4-8GHz). Transmission data (S21) was taken for this measurement.

Results and Discussion

Fig. 1 shows the data from XRD pattern of GdIG. It shows no secondary phase formation where all peaks are matched with the reference code pattern of JCPDS 01-074-1361. This proves the ability of the sample to crystallize at lower sintering temperature as compared to that of reported by [5], where the sufficient heating treatment given to form fully garnet structure is above 1200 ^oC. This might be due to the mechanical alloying that has initiated the initial stage of crystallization with a very high impact collision between balls and powder that allows the repetitive alloying, cold welding, and fracturing mechanism, thus formed very high reactivity of particles with large surface-to-volume ratio. This occurrence will lead to very high Gibb's free energy that lowers the activation energy and consequently reduces the temperature required for single phase formation of GdIG [6].

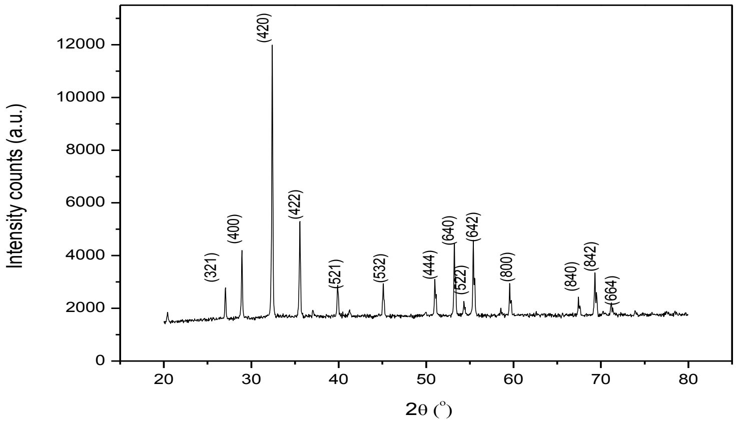
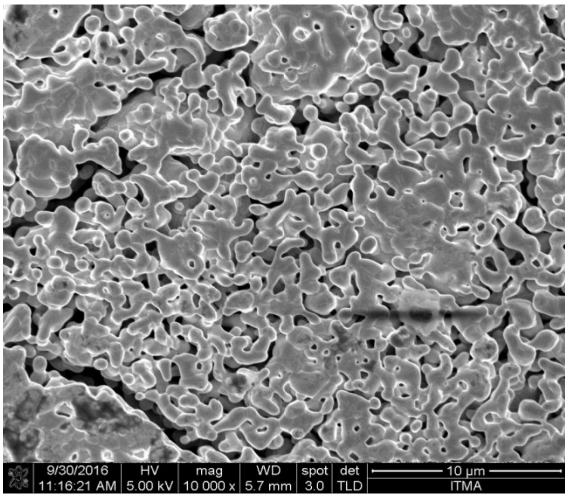


Fig. 1: XRD pattern of single phase 3 hour milled GdIG, sintered at 1200 ^oC

Fig. 2 (a) and 2 (b) show the micrograph of grains and the histogram of grain size distribution respectively. Mechanical alloying leads to agglomeration between the particles due to high reactivity of large surface area besides the contribution from cold welding and alloying effect. Hence, the post-sintering grains observed in Figure 2 (a) are also agglomerated. It also can be observed that the size of pores is still big in the sample and grain boundaries have appeared clearly. Pores and grain boundaries are non-magnetic inclusion that might affect the magnetic and microwave properties that will be discussed later. The histogram shown in Figure 2 (b) signifies that the grains are not well distributed for the high energy impact between balls and powder particles was inconsistent in the process. The average grain size measured is ~0.84 µm.

(a)



(b)

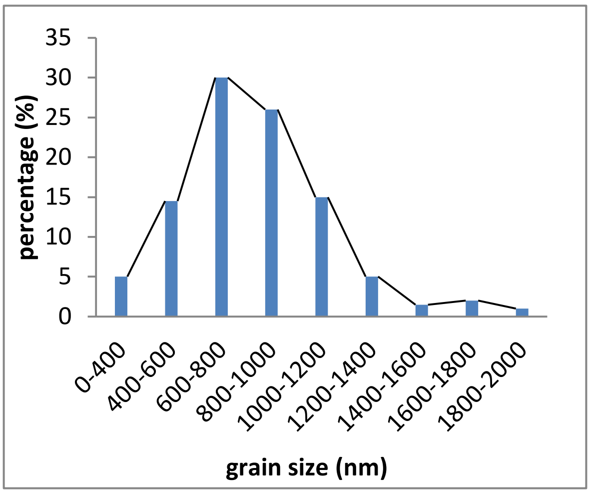


Fig. 2 (a): FESEM image of GdIG microstructure, (b): histogram of grain size distribution

Results of complex permeability are shown in Fig. 3 (a) and (b). From the figure, it has been identified that the initial permeability of GdIG is rather low. Permeability is mainly influenced by either spin rotation or domain wall movement. At low frequency, domain wall movement effect gives greater contribution to the value of permeability. As a result of sintering process, grain growth will eventually improve the size of grains, reduced the grain boundary and increase the crystallinity of the sample. Larger grain size results in easier movement of domain wall, thus increased the value of real permeability. However, grain boundaries and pores alongside the grains would act as impediment which hinder the movement of domain wall. Therefore, the existence of both hindrances might lower the permeability of a material [7]. It can be observed in Figure 2 (a) where there are great size of pores and grain boundaries. The loss factor can be fractionated into 5 types; dielectric loss, hysteresis loss, domain wall resonance, ferromagnetic resonance, and eddy current loss [8]. Since ferrite is a high resistive material, the effect of eddy current and the spin rotation contribution can be neglected due to low frequency range of energy applied (10MHz – 1 GHz). In the case of GdIG loss factor, domain wall resonance and hysteresis loss dominate the effect.

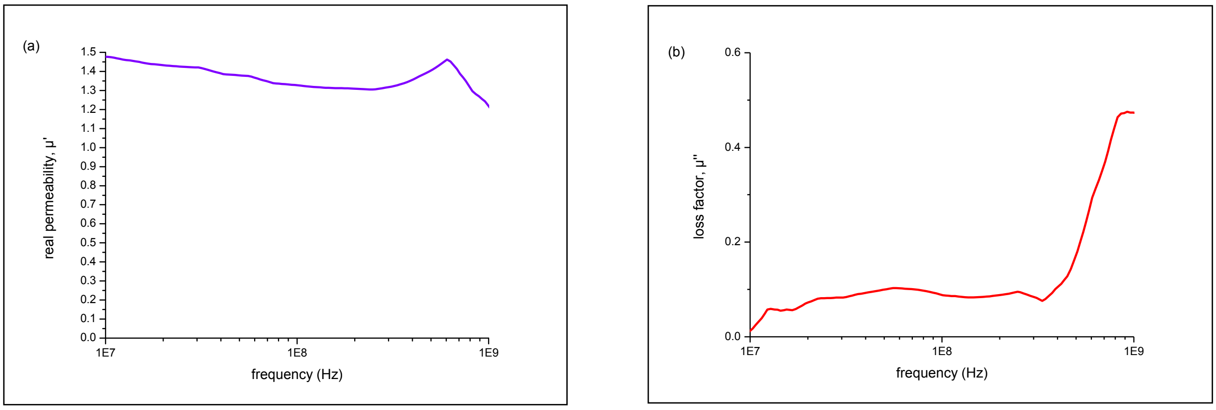


Fig. 3 (a): Graph of real permeability and (b) loss factor of GdIG in the frequency range of 10MHz-1GHz

Fig. 4 showed a reflection/transmission loss of the GdIG sample measured using VNA at C-band frequency range. Ferromagnetic resonance (FMR) linewidth, ΔH was calculated from the transmission (S_{21}) data acquired using the following equation, Eq. 2:

$$\Delta H = \frac{\Delta\omega}{\gamma} \tag{2}$$

where $\Delta\omega$ is angular frequency bandwidth and the frequency bandwidth was taken from the full wave half maximum (FWHM) of S_{21}. γ is gyromagnetic ratio where the value is $1.76 \times 10^{11} s^{-1}T^{-1}$. Transmission loss depicts on how much energy can be transmitted through GdIG. The lower the transmission loss, the higher the energy transmitted. The value of ΔH is predominantly contributed from extrinsic properties such as porosity and anisotropy of the material itself [9, 10]. The value of ΔH obtained from the calculation is 14.53 Oe which is a lot smaller than that of the value required for low loss microwave material (100 Oe) [5]. This is due to the small anisotropy constant of GdIG [11] which means the bigger the value of anisotropy constant of a material, the bigger the value of ΔH will be obtained. GdIG received sufficiently high energy that spin resonance dominating at high frequency besides the effect from eddy current that cannot be neglected at high frequency range. The microstructure of GdIG also plays an important role that affect the microwave property. Linewidth also can be broadened by virtue of various crystal orientations. Grain boundaries, non-magnetic inclusions and inhomogeneous regions are the other factors inclusive [12].

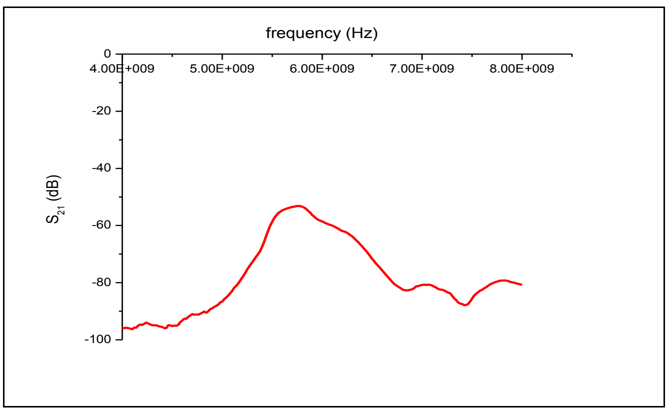


Fig. 4: VNA data of transmission loss (S_{21}) of GdIG

Conclusion

GdIG was successfully prepared via the mechanical alloying technique. It is proven that the magnetic and microwave properties are closely related to the microstructure of GdIG. Although the permeability shows low value at low frequency range, low microwave loss result showed that GdIG is a suitable material to be used in high frequency applications.

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