Yttrium iron garnet ceramic prepared from microwave-induced combustion

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Abstract Yttrium iron garnet (YIG) nano-powders were successfully synthesized by microwave-induced combustion process. The as-prepared YIG powders were annealed at different temperature and investigated by XRD, SEM, and VSM. The as-received YIG powder shows the formation of garnet structure with saturation magnetization (M_s) of 12 emu/g, whereas upon annealing at 825 °C for 2 h, the saturation magnetization increases to 27 emu/g. The asreceived yttrium iron garnet powders annealed at several temperatures revealed that the particle size ranged from 65 to 90 nm. Moreover, the linewidth of magnetic resonance peak (Δ H), complex permeability, and loss tangent of sintered YIG ceramic are also examined in this study.

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1 Introduction

Yttrium iron garnet (YIG) was extensively studied owing to its interesting physical properties. For example, this material possesses the highest quality factor in microwave regime, and the smallest linewidth in magnetic resonance, among the magnetic materials [1, 2]. They are important components for the electronics products, such as circulators, oscillators, and phase shifter. The conventional way of producing these materials is by the solid state reaction of with oxide/carbonate and calcined at high temperature (≥ 1200 °C). The solid-state reaction method has some inherent disadvantages: such as (1) chemical inhomogeneity, (2) coarser particle size, and (3) introduction of impurities during ball milling [3].

In the present work, we attempted a new method called microwave-induced combustion synthesis process to produce yttrium iron garnet nano-powders. The combustion synthesis process is to dissolve metal nitrates and urea in water [4], and then to heat the solution in a microwaveoven. The whole process takes only a few minutes to yield powders of YIG. The result in the formation of single phase YIG powder after synthesizing in the microwaveoven with saturation magnetization of 12.5 emu/g, whereas upon annealing at 825 °C for 2 h, the saturation magnetization (M_s) increased to 27.6 emu/g. YIG powders annealed at various temperatures were further characterized by XRD, SEM, and VSM measurements and the results are described in this paper. Additionally, the complex permeability, loss tangent, and the magnetic resonance property of sintered YIG ceramic are also described in this paper.

2 Experimental procedures

2.1 Sample preparation

2.1.1 Preparation of YIG powders

The synthesis process involved the combustion of redox mixtures, in which metal nitrate acted as an oxidizing reactant and urea as a reducing one. The initial composition of the solution was based on the total oxidizing and reducing valences of the oxidizer and the fuel using the concepts of the propellant chemistry [5]. Yttrium nitrate [Y (NO₃)₃·6H₂O], ferric nitrate[Fe(NO₃)₃·9H₂O], and urea [CO(NH₂)₂] were dissolved in a minimum quantity of water were set in a crucible. The crucible containing the solution was introduced into a microwave oven (CEM, MDS 81D, 650W). Initially, the solution boils and undergoes dehydration followed by decomposition with the evolution of large amount of gases (N₂, NH₃ and HNCO). After the solution reaches the point of spontaneous combustion, it begins burning and releases lots of heat, vaporizes all the solution instantly and becomes a solid burning at high temperature. The entire combustion process produces YIG powders in a microwave-oven takes only 15 min. The as-prepared YIG powders were annealed at different temperatures from 750 to 850 °C for 2 h.

2.1.2 Preparation of sintered YIG ceramic

The as-prepared YIG powder annealed at 825 °C for 2 h, and then these annealed YIG powder was granulated, pressed into toroidal specimens under a uniaxial pressure of 1000 kgf/cm². Toroidal specimens were sintered in air from room temperature to 1325, 1350, and 1375 °C at a heating rate of 3 °C, and annealed at these temperatures for 4 h, respectively.

2.2 Characterization

A computerized X-ray powder diffractometer (XRD) with Cu K_{α} radiation was used to identify the crystalline phase. The magnetic properties of as-prepared YIG powders annealed at different temperatures were measured by a vibrating sample magnetometer (VSM) at room temperature. The particle morphological features were imaged by scanning electron microscopy (SEM). The complex permeability and loss tangent of sintered YIG ceramic were measured on an HP E4991A impedances analyzer from

1 MHz to 1 GHz. The magnetic resonance properties measured by electron paramagnetic resonance (EPR) spectrometer were used to elevate the Q-factor in microwave regime.

3 Results and discussion

The X-ray diffraction patterns of the as-received YIG powder and as-received specimen annealed at different temperatures are shown in Fig. 1. It is evident that the annealed as-received YIG powders contain only the garnet phase. Y₃Fe₅O₁₂ is formed all the peaks in the pattern match well with JCPDS card. No obvious difference can be found among the XRD patterns of samples annealed from 750 to 850 °C. The magnetization measurements for the asreceived specimen and the variously annealed specimens were carried out by using VSM at room temperature with an applied magnetic field of 12 kOe to reach saturation values, which results are shown in Fig. 2. YIG powders prepared by combustion route and annealed at 825 °C for 2 h exhibits an optimum M_s of 27.6 emu/g. It shows that the $M_{\rm s}$ increases steadily with increasing annealing temperature from 750 to 825 °C, as a result of increasing good crystallinity. Further increasing annealing temperature above 825 °C led to a fall in M_s to 24.7 emu/g at 850 °C. The Hc demonstrates an initial rise from 750 to 825 °C, and it peaks at 825 °C. The as-received YIG powder annealed at 825 °C exhibits a largest Hc than the other annealing temperatures. Figure 3 shows the variation of complex permeability of YIG sintered at various temperatures. Real part of complex permeability (μ') is influenced by many factors such as large, homogenous grains, chemical homogenous, and no second phase [6, 7]. Therefore, YIG specimen sintered at 1350 °C displays the highest permeability at frequency less



Fig. 1 XRD powder pattern of as-received and annealed YIG at different temperatures



Fig. 2 Variation of magnetization with the applied magnetic field for YIG samples annealed at different temperatures

than 180 MHz might be attributed to a larger achieved grain size compared to the specimens sintered at different temperatures. The relaxation seems to occur significant in the range of 1–50 MHz in all sintering condition. Imaginary part of complex permeability (μ^{ll}) is related to many factors



Fig. 3 Complex permeability of YIG ceramic sintered at different temperatures with variation of frequency



Fig. 4 Loss tangent of YIG ceramic sintered at different temperatures with variation of frequency

such as rotation of magnetization and domain-wall displacement, which shows two resonances approximately at 180 and 880 MHz, respectively. The fraction occupied by the domain-wall part will increase if the grain size is large; this will increase eddy current and hysteresis loss [8]. YIG specimen sintered at 1350 °C reveals the highest value of imaginary part of complex permeability at frequency less than 600 MHz, as frequency above 600 MHz, YIG specimen sintered at 1375 °C shows the highest value. Figure 4 depicts the frequency dispersion of loss tangent for YIG specimens sintered at various temperatures. It indicates that YIG specimens possess two maximum approximately at 850 and 900 MHz, respectively in all specimens. Furthermore, YIG specimens sintered at 1350 °C reveals the lowest value of loss tangent. This suggests that appropriate sintering temperature is good for YIG ceramic magnetic properties at high frequency.

The linewidth of resonance peaks (Δ H), depends on a variety of factor. Anisotropy field effects can be expressed in



Fig. 5 Magnetic resonance spectra for YIG ceramic sintered at 1350 °C for 4 h, showing that the linewidth of the resonance peak (Δ H) of 53 Oe

term of a damping constant leading to a contribution to ΔH . If the dc magnetic field is not strong enough to saturate the sample, the total field in each domain is different and ΔH increases. In polycrystalline species this effect is more serious, because interactions between grains result in larger variations in the total field. Local perturbations in total field produced by porosity lead to a linear increase in linewidth. Figure 5 displays the linewidth of YIG ceramic sintered at 1350 °C for 4 h, revealing that the ΔH of 53 Oe.

4 Conclusions

Using yttrium nitrate, ferric nitrate, and urea as the starting materials, fine YIG powders have been synthesized successfully by microwave-induced combustion process. The saturation magnetization of YIG powder can be significantly improved by an appropriate annealing temperature, while the saturation magnetization peaks at 27 emu/g

for the annealing temperature at 825 °C for 2 h. For YIG ceramic, the specimens sintered at 1350 °C revealed the narrow linewidth of the resonance peak (Δ H) of 53 Oe.

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