The Effect of Both B_2O_3 and CuO Additions on Magnetic and Microwave Properties of YIG

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Abstract— The structure and magnetic and microwave properties of polycrystalline YIG, prepared by a conventional ceramic method with a small addition of both B_2O_3 and CuO as a sintering agent, were studied using X-ray diffractometry, magnetometry and ferromagnetic resonance experiments. Samples sintered with the sintering agent consisted of mainly a cubic garnet structure and $CuOFe_2O_3$ particles formed only on free surface. Saturation magnetization of the polycrystalline garnets was about 27 emu/g at room temperature and almost independent of the amount of B_2O_3 + CuO addition in the range of 0.0 -2.0 wt%. Ferromagnetic resonance linewidth measured at 9.4 GHz decreased from about 400 to 58 Oe with increasing B_2O_3 and CuO content from 0.0 to 1.5 wt% due to increased sinterability. We can reduce sintering temperature by about 150 K by adding small amount of B_2O_3 + CuO.

I. INTRODUCTION

YIG (Yttrium iron garnet) has been widely used because of its small resonance linewidth in many devices such as isolator, circulator and gyrators [1-4]. Pure and substituted YIGs have played a central role in research and applications concerning ferromagnetic resonance. The saturation magnetization, resonance linewidth, microwave permittivity and permeability are properties of fundamental importance in determining the performance of the devices in the microwave region [5,6]. Several types of substituted garnets for high frequency applications were developed to tailor and to improve their magnetic and microwave properties. Partial substitution of both yttrium and iron by other ions can change magnetic and microwave properties. Addition of dopants affects the intrinsic properties such as magnetization, anisotropy and electrical resistivity in MnZn ferrite due to liquid phase formation during sintering and to modification of grain boundary chemistry [7]. Addition of dopants may affect not only magnetic and microwave properties of YIG but also sintering behaviour. Reducing sintering temperature may offer several economic advantages especially in the commercial application of YIG. In this study we have added small amount of both B_2O_3 and CuO to calcinated YIG powder with the intention of reduce sintering temperature and improving microwave properties.

II. EXPERIMENTS

Polycrystalline YIG specimens were prepared by a conventional ceramic method. The raw materials, yttrium oxide Y_2O_3 and ferric oxide $\alpha-Fe_2O_3$ were weighed to form a stoichiometric YIG and then thoroughly mixed by using a ball mill. Calcination of the mixture was performed at 1473 K for 2 hrs in the atmosphere. Small amount of both B_2O_3 and CuO was added to the calcinated powder and then homogeneous mixture was obtained by using a ball mill. Total amount of B_2O_3 and CuO varied from 0.0 to 2.0 wt%. The powders mixed with polyvinyl alcohol (PVA) as a binder were cold pressed at 150 MPa to make coins. Each coin was then sintered at temperatures of 1523, 1573, and 1623 K. For the structural characterization of the resulting samples, X-ray diffraction (XRD) experiments were performed with monochromatic Cu-K α radiation on both free and polished surfaces of sintered samples. The densities of the prepared garnet samples were measured by using Archimedes' method. The microstructure of the samples was observed by scanning electron microscopy (SEM). Also microcompositional analysis was performed by Energy dispersive X-ray spectroscopy (EDX). The temperature dependence of the saturation magnetization of the samples was measured by vibrating sample magnetometer (VSM) from room temperature to 600 K with an external field of 5 kOe. Also ferromagnetic resonance experiment was carried out to measure ferromagnetic resonance linewidth (ΔH) by using spherical samples of about 0.5 mm in diameter at room temperature and a frequency of 9.44 GHz.

III. RESULT AND DISCUSSION

XRD experiments were carried out on both free and polished surfaces of sintered specimens.

Fig. 1. XRD traces obtained from (a) free surface and (b) polished surface of specimen sintered at 1573 K with 1.5 wt% of B_2O_3 and CuO.

Figs. 1 (a) and (b) show, respectively, XRD traces obtained from the free surface and polished surface of specimen, sintered at 1573 K with 1.5 wt% of B_2O_3 and CuO. As can be seen from the analysis, the XRD trace from the free surface consisted of diffraction peaks corresponding to both a Cu ferrite ($CuOFe_2O_3$) phase and a cubic garnet phase. However, the XRD trace obtained from the polished surface of the specimen consisted of diffraction peaks corresponding to only YIG phase as marked by solid circle (●). The XRD results indicate that Cu ferrite only formed on the free surface of the specimen. This may explain the high intensities of diffraction peaks Cu ferrite in Fig. 1(a), even though small amount of CuO was added. During sintering liquid phase formed at grain boundaries due to the added sintering agents with low melting points. The liquid phase can accelerate densification process and the liquid phase seems to be pushed out to the free surface as densification proceeds. The densities of specimens, sintered at 1523 K with different CuO and B_2O_3

contents, were measured without polishing. The densities of specimens varied from about 4.88 to about 5.08 g/cm³ with increasing sintering agent from 0.5 to 1.0 wt% of CuO and B_2O_3 . Further increase in sintering agent did not give any effect on the density of the specimen. The density of YIG specimen, sintered at 1673 K without sintering agent, was about 4.96 g/cm³. The results clearly indicate that small addition of B_2O_3 and CuO improved the sinterability of the YIG specimen, and that the addition of CuO and B_2O_3 can lower sintering temperature, which may offer many advantages in commercialization of the process.

Figs. 2 (a) and (b) show SEM microstructures taken from the free surfaces of YIG specimens, sintered at 1523 K, with CuO and B_2O_3 content of 0.5 wt% and 2.0 wt% respectively. Grain size of the specimen increased from about 5 μ m to 50 μ m with increasing CuO and B_2O_3 content from 0.5 to 2.0 wt% without forming void. Also faceted second phase was observed as shown in fig.2. The increase in grain size may

Fig. 2. SEM microstructure taken from free surface of YIG specimens, sintered at 1523 K, (a) with 0.5 wt% and (b) 2.0 wt% of B_2O_3 and CuO.

arise from increased liquid fraction during the sintering process and is in agreement with increased sinterability.

Fig.3. EDX spectra taken (a) from matrix and (b) from faceted particle in YIG specimen sintered at 1523 K, with 2.0 wt% of B_2O_3 and CuO.

Figs. 3 (a) and (b) show EDX spectra taken from a matrix and a faceted particle respectively in YIG specimen sintered at 1523 K with B_2O_3 and CuO of 2.0 wt%. For EDX analysis specimens were coated with Au. Therefore a weak peak from Au is expected to appear. The EDX spectrum from a matrix consisted of peaks corresponding to yttrium, iron and oxygen.

However, EDX spectrum from a faceted particle consisted of peaks corresponding to copper, iron and oxygen. Considering the strong diffraction peaks corresponding to CuOFe_2O_3 as shown in Fig. 1(a) and EDX results as shown in Fig. 3(b), the faceted particles formed on the free surface as shown in Fig. 2 are likely to be CuOFe_2O_3 .

Fig. 4. Temperature dependences of M_s of pure YIG sample sintered at 1673 K (○) and YIG sintered at 1523 K with 2.0 wt% of B_2O_3 and CuO (●).

Fig. 4 shows the temperature dependences of M_s of pure YIG sintered at 1673 K and YIG specimen sintered at 1523 K with 2.0 wt% of B_2O_3 and CuO. The Curie temperature of the specimens was about 570 K, which was almost independent of the content of B_2O_3 and CuO. Also we can see that the saturation magnetization was not affected by the small addition of B_2O_3 and CuO. These results seem to be in agreement with the XRD experiment. In order to measure M_s , the sintered specimens were ground to small spherical shapes and therefore CuOFe_2O_3 particles were all removed from the specimen during grinding.

Fig. 5 shows a variation of ΔH of the specimens, sintered at 1523 K, as a function of B_2O_3 and CuO content. Also ΔH s of YIG specimens sintered without sintering agent at 1673 K for 2 hrs and for 8 hrs were included for comparison. ΔH value of pure YIG sample, sintered without B_2O_3 and CuO at 1523 K showed about 400 Oe due to lots of pores in the specimen. However, a slight addition of B_2O_3 and CuO improved sinterability. With increasing the content of B_2O_3 and CuO, ΔH value of the specimens decreased to 88 Oe at 0.5 wt% of B_2O_3 and CuO, reached a minimum value of 58 Oe at 1.0 and 1.5 wt%, and then increased slightly to 65 Oe at 2.0 wt% of B_2O_3 and CuO. Pure YIG specimen sintered at 1673 K for 2 and 8 hrs showed ΔH values of 65 and 55 Oe, respectively. The minimum ΔH value obtained from the specimens sintered at 1523 K with 1-1.5 wt% of B_2O_3 and CuO was comparable with the ΔH value of pure YIG sample sintered at

1673 K for 8 hrs. The specimen sintered at 1523 K with 1.0 or 1.5 wt% B_2O_3 and CuO showed lower ΔH than pure YIG specimen sintered 1673 K during 2 hrs as shown in Fig. 5. Finally, sintering temperature could be reduced by about 150 K by adding 1.0 and 1.5 wt% B_2O_3 and CuO without changing magnetic properties of YIG while simultaneously improving its microwave property.

Fig. 5. A variation of ΔH of YIG specimens sintered at 1523 K, with B_2O_3 and CuO content. Also ΔH values of pure YIG specimens sintered at 1673 K were included for comparison.

IV. CONCLUSIONS

We have added small amount of B_2O_3 and CuO to calcinated YIG powder with the intention of reducing the sintering temperature. CuOFe_2O_3 particles were formed only on the free surface of the specimens, sintered with small addition of B_2O_3 and CuO. Sintering temperature can be reduced by about 150 K by adding 1.0 and 1.5 wt% B_2O_3 and CuO without any change in saturation magnetization and Curie temperature. The specimen prepared with B_2O_3 and CuO showed improved microwave property possibly due to reduced porosity.

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