

# The effect of SrTiO<sub>3</sub> addition on the magnetic and microwave properties of yttrium iron garnet

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**Abstract** The effect of SrTiO<sub>3</sub> addition on the microstructure and magnetic properties of yttrium iron garnet prepared by a conventional ceramic method was investigated using scanning electron microscopy, X-ray diffractometry, vibrating sample magnetometry, and ferrimagnetic resonance experiments. A YIG specimen sintered at 1693 K with 0.2 mol% SrTiO<sub>3</sub> addition showed above 98% densification of theoretical density without changing in magnetic properties of YIG. A YIG specimen sintered at 1693 K with 0.2 mol% SrTiO<sub>3</sub> showed ferrimagnetic resonance linewidth  $\Delta H$  of about 80 Oe.

## Introduction

Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> (YIG), a ferrimagnetic materials known as microwave ferrite, has widely applied in passive microwave devices since its discovery in 1956 [1–3]. Due to their microwave properties, YIG and its substituted derivatives are of primary interest for use in the conceptions of microwave equipment (isolators, circulators, dephasers). Recently, YIG has been prepared by a variety of methods such as liquid phase epitaxy (LPE), radio frequency sputtering, pyrolysis, metallo-organic decomposition (MOD), and chemical vapor decomposition (CVD) in film [4–7]. In most cases, the materials used are, however, sintered

polycrystalline ceramics practically. In order that the materials developed possess the required magnetic characteristics they must be pure, homogeneous and have a density very close to the theoretical value. A high sintered density and uniform microstructure are desirable to minimize the magnetic loss at this high frequency [8–10].

In the present study, we investigated the effect of small amounts of SrTiO<sub>3</sub> addition as a sintering additive on various properties of YIG prepared by conventional ceramic method. It is expected that SrTiO<sub>3</sub> addition—Sr ions are similar to Y ions in ionic radius and Ti ions are used for electrical neutrality—contributes to various properties of YIG.

## Experimental procedure

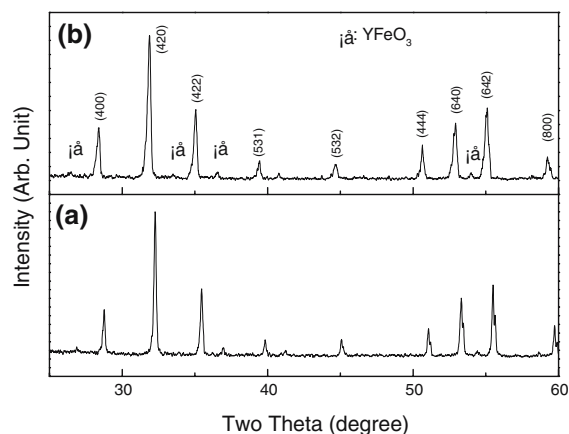
A conventional ceramic method was adopted for synthesizing the polycrystalline YIG specimens. The raw materials, yttrium oxide Y<sub>2</sub>O<sub>3</sub> (purity 99.9%) and ferric oxide Fe<sub>2</sub>O<sub>3</sub> (purity 99.9%) were weighted to form stoichiometric YIG and then thoroughly wet mixed using a ball mill. The mixture was dried, calcined at 1573 K for 6 h in air, then crushed into powder. An appropriate amount of SrTiO<sub>3</sub> (purity 99.9%) was added to the calcinated powder and then homogeneous mixture was obtained using a ball mill. The amount of SrTiO<sub>3</sub> addition was varied from 0.0 to 0.3 mol%. The mixed powders were pressed into disk-shaped samples and cold-pressed at 200 MPa. The disc was then sintered at various temperatures of 1593 and 1693 K. For structural characterization of the resulting samples, X-ray diffraction (D/MAX-11A, Rigaku) was performed with monochromatic CuK $\alpha$  radiation. The bulk

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density and shrinkage of the prepared garnet sample were measured using ASTM (American Society for Testing and Materials) standards. Microstructure and grain size were estimated by scanning electron microscopy (JEOL 4500, JEOL). The saturation magnetization ( $M_s$ ) and coercive force ( $H_c$ ) of the samples were measured using a vibrating sample magnetometer (7300 VSM, Lake Shore) with an external field of 2.5 kOe. In addition, ferrimagnetic resonance experiments were carried out to measure the ferrimagnetic resonance (JES-TE300, JEOL) linewidth ( $\Delta H$ ) using a almost spherical sample of about 1.0 mm in diameter at room temperature.

## Results and discussion

Figure 1(a) and (b) show XRD patterns obtained from YIG samples sintered at 1693 K with 0.0 and 0.2 mol% of  $\text{SrTiO}_3$ , respectively. Both XRD patterns consisted of diffraction peaks corresponding to the cubic YIG phase. Also a very weak peak corresponding to orthoferrite  $\text{YFeO}_3$  phase appeared. The intensity of the orthoferrite phase peak increased slightly with increasing  $\text{SrTiO}_3$  content. The formation of the orthoferrite phase indicated that partial substitution of Y ions with Sr ions occurred during sintering. However, the intensity of the orthoferrite phase peak was relatively very low compared to the main peak of pure YIG peak (420) indicating that the amount of Sr ion substitution was small. The lattice constant ( $a_0$ ) of the YIG sample, calculated from diffraction peaks in Fig. 1, increased slightly from about 12.402 to 12.457 Å with increasing  $\text{SrTiO}_3$  content from 0.0 to 0.3 mol% and follows Vegard's rule approximately. This can be expected in view of the fact that the radius of 0.95 Å for and Y ion is smaller than that of 1.13 Å for a Sr ion.

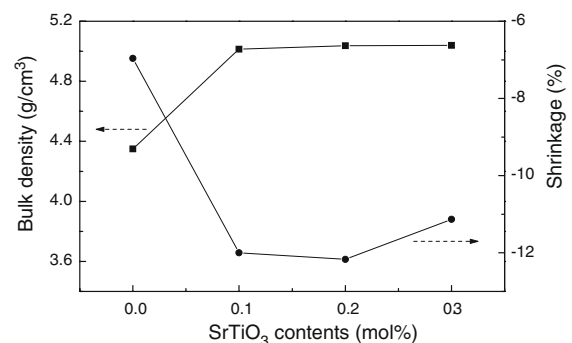


**Fig. 1** X-ray diffraction patterns of YIG specimens sintered at 1693 K with  $\text{SrTiO}_3$  contents of (a) 0.0 mol% and (b) 0.2 mol%

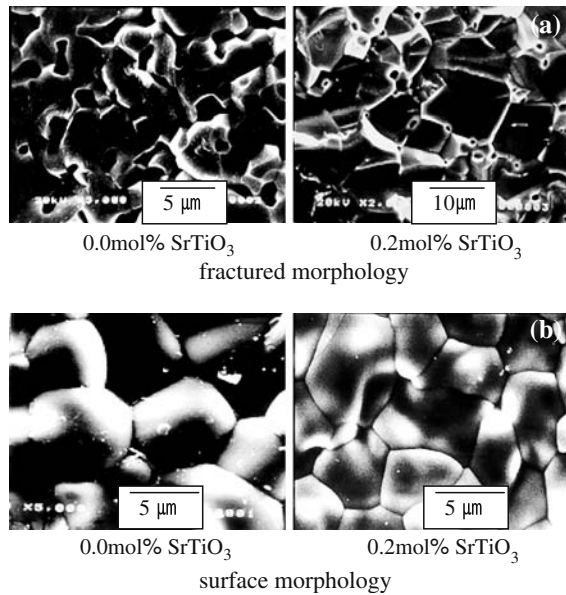
Figure 2 indicates that the bulk densities and shrinkages of YIG specimens prepared at 1693 K with different amounts of  $\text{SrTiO}_3$  addition were measured. The shrinkage of YIG increased from 7% at pure YIG sintered at 1693 K and reached a maximum value of 12.2% at 0.2 mol%  $\text{SrTiO}_3$ . The bulk density of YIG sintered at 1693 K increased from 4.96 at pure YIG and reached a maximum value of 5.07 at 0.2 mol%  $\text{SrTiO}_3$  and then decreased slightly to 5.01  $\text{g/cm}^3$  at 0.3 mol%  $\text{SrTiO}_3$ . This showed that our samples had above 98% densification of theoretical density 5.17  $\text{g/cm}^3$  of YIG from X-ray density. From the above results, it would be suggested that 0.2 mol%  $\text{SrTiO}_3$  addition improves sinterability of YIG specimens.

Figure 3 shows the SEM microstructure of YIG specimens sintered at 1693 K, with  $\text{SrTiO}_3$  content of 0.0 and 0.2 mol%, respectively. Figure 3(b) indicated that surface morphology of YIG with both 0.0 and 0.2 mol%  $\text{SrTiO}_3$  addition had grain structure with no void. However, Fig. 3(a) showed that YIG specimen sintered with 0.2 mol%  $\text{SrTiO}_3$  addition was more dense than without  $\text{SrTiO}_3$  addition. This would be a good agreement with above bulk density and shrinkage data. The grain size of the specimens increased from about 2.5 to 5.4  $\mu\text{m}$  with increasing sintering temperature but seemed not to be affected by  $\text{SrTiO}_3$  content.

The saturation magnetization of YIG samples prepared at 1693 K with different  $\text{SrTiO}_3$  contents from 0.0 to 0.3 mol% was measured VSM with an external field of 2.5 kOe. Increasing the amount of  $\text{SrTiO}_3$  from 0.0 to 0.3 mol% resulted in the room temperature magnetization decreasing slightly from 28.5 to 28 emu/g. The decrease in magnetization in our sample seemed to result from the formation of the orthoferrite phase at higher  $\text{SrTiO}_3$  contents. This result is similar to the other researcher's study [11]. The coercive force of the specimens decreased from about 10.5 to 8.3 Oe with increasing sintering temperature but seemed not to be affected by  $\text{SrTiO}_3$  content. This phenomena is



**Fig. 2** Bulk densities and shrinkages of YIG sintered at 1693 K with  $\text{SrTiO}_3$  contents



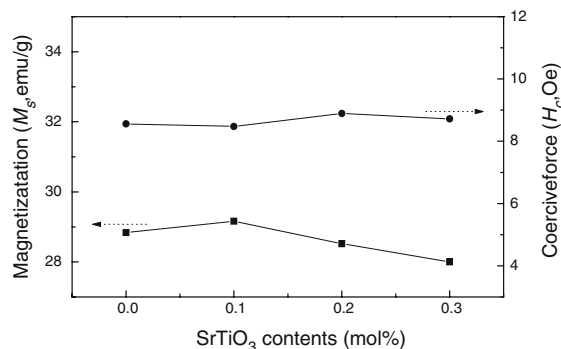
**Fig. 3** Scanning electron micrographs of (a) fractured morphology and (b) surface morphology of YIG specimens sintered at 1693 K with SrTiO<sub>3</sub> contents of 0.0 and 0.2 mol%, respectively

consistent with that coercive force, in general, is the very sensitive with grain size [12] (Fig. 4).

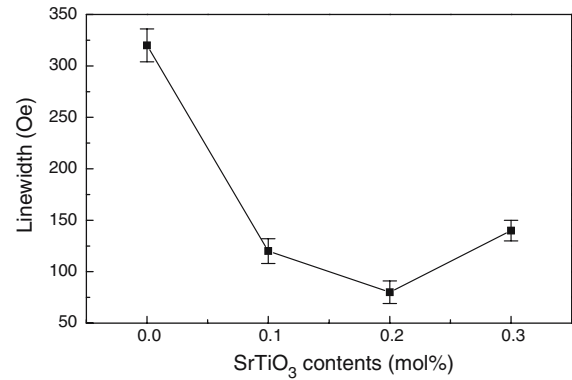
Figure 5 shows the variation of  $\Delta H$  with SrTiO<sub>3</sub> content for YIG specimens sintered at 1693 K.  $\Delta H$  values of the YIG samples decreased from about 330 Oe, reaching a minimum value of 80 Oe at 0.2 mol% SrTiO<sub>3</sub> and then slightly increased with increasing SrTiO<sub>3</sub>. The standard technique to study microwave losses in ferrites is to measure ferrimagnetic resonance (FMR) linewidth. FMR linewidth ( $\Delta H$ ) in polycrystalline ferrites can separated in different contributions [13]:

$$\Delta H = |K_1|/M_s + (8/\pi\sqrt{3})(4\pi M_s)\{p_{\text{eff}}/(1 + p_{\text{eff}})\} + \Delta H_i$$

where  $K_1$  is the first-order anisotropy constant,  $M_s$  the saturation magnetization,  $p_{\text{eff}}$  the fraction of effective



**Fig. 4** Magnetizations ( $M_s$ ) and coercive forces ( $H_c$ ) of YIG sintered at 1693 K with SrTiO<sub>3</sub> contents



**Fig. 5** Ferrimagnetic resonance linewidths ( $\Delta H$ ) of YIG specimens sintered at 1693 K with SrTiO<sub>3</sub> contents

nonmagnetic volume (demagnetizing effects of porosity including second phase), and  $\Delta H_i$  the linewidth of the corresponding single crystal sample, its value is usually much smaller than those of the corresponding polycrystalline materials, can be neglected. Also effect of  $M_s$  is negligible because  $M_s$  value is not greatly changed in our samples. A pure YIG specimen sintered at 1693 K showed a large  $\Delta H$  value due to the presence of pores in the specimen as described previously. By adding small amounts of SrTiO<sub>3</sub>,  $\Delta H$  of YIG specimens sintered at 1693 K showed a large decrease due to the increased sinterability and lower porosity (the effect of  $p_{\text{eff}}$  decreasing). The slight increase in  $\Delta H$  at 0.3 mol% SrTiO<sub>3</sub> seems to be related with the increasing of the orthoferrite phase YFeO<sub>3</sub>, which acts as a second phase in above equation, as shown in Fig. 1.

## Conclusion

In this study, the lattice constant of the YIG sample increased slightly from about 12.402 to 12.457 Å with increasing SrTiO<sub>3</sub> content from 0.0 to 0.3 mol% because partial substitution of Y ions with Sr ions occurred during sintering. The bulk density of YIG increased from 4.96 at pure YIG and reached a maximum value of 5.07 at 0.2 mol% SrTiO<sub>3</sub>, which showed above 98% densification of theoretical density.  $\Delta H$  values of the YIG samples decreased from about 330 Oe, reaching a minimum value of 80 Oe at 0.2 mol% SrTiO<sub>3</sub>. From the result, it seems that YIG sintered at 1693 K with 0.2 mol% SrTiO<sub>3</sub> addition have a good properties by improvement of sinterability without variation of magnetic properties.

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