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Effect of La_2O_3 doping on the tunable and dielectric properties of BST/MgO composite for microwave tunable application

Rui-Hong Liang *, Xian-Lin Dong, Ying Chen, Fei Cao, Yong-Ling Wang

Shanghai Institute of Ceramics, Chinese Academy of Sciences, 1295 Dingxi Rd., Shanghai 200050, PR China

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Abstract

Undoped and La_2O_3doped (0.5, 1.0, 2.0, 3.0 wt.%) $Ba_{0.55}Sr_{0.45}TiO_3$/MgO composites were prepared by traditional ceramic processing and their structural, surface morphological, tunable properties and their dielectric properties at low frequency and microwave frequency were systemically examined. The result shows that La_2O_3 dopant has a strong effect on the average grain size. The La_2O_3 doped samples have lower temperature coefficient of capacitance than the undoped. The 0.5 wt.% La_2O_3 doped sample has a little higher tunability than the undoped and the tunability of other doping concentration samples is lower as compared to the undoped. The addition of La_2O_3 decreases the dielectric constant and increases quality factor ($Q \times f$) at microwave frequency. The 0.5 wt.% La_2O_3 doped samples have the best properties among these samples and have a higher tunability, lower dielectric constant and lower dielectric loss tangent at microwave frequency and these properties are very beneficial to the development of the tunable devices application.

Keywords: Ferroelectric–nonferroelectric oxide composites; Tunable property; Microwave dielectric property

1. Introduction

Present phased-array antennas are constructed using ferrite phase shifting elements. These elements are ferromagnetic and current driven, where the phase shift is caused by a change in the permeability of the material. The performance of this type phase shifter is good, however they are very costly, large, and heavy. In order to make these devices more practical, smaller and can be used in higher frequency, better materials must be developed. The strong dependence of dielectric constant on electric field of ferroelectric materials with perovskite structure have made them prospective candidates for microwave tunable applications, such as phase shifters, tunable filters, steerable antennas. The desired material properties are quite well known: low dielectric constant, a high tunability, low dielectric losses at microwave frequency and low temperature coefficient of the capacitance are required. Recently, the common studies in this field are focused on $Ba_xSr_{1-x}TiO_3$ and nonferroelectric oxides composite (BSTO) system [1–3]. In this paper, we report an investigation of the structural, surface morphological properties, tunability and dielectric properties at low frequency and microwave frequency of $Ba_{0.55}Sr_{0.45}TiO_3$/MgO composite system as a function of La_2O_3 dopant concentration from 0 to 3.0 wt.% and studied whether the addition of dopant can optimize the properties of BST/MgO system.

2. Experiments

Undoped and La_2O_3 doped (0.5, 1.0, 2.0, 3.0 wt.%) $Ba_{0.55}Sr_{0.45}TiO_3$/MgO composites were prepared by traditional ceramic processing. The weight ratio of $Ba_{0.55}Sr_{0.45}TiO_3$ and MgO is 1:1. During the first stage the starting powder ($Ba_{0.55}Sr_{0.45}TiO_3$) was synthesized by mixing $BaCO_3$, $SrCO_3$ and TiO_2 in ethanol on a ball mill for 24 h and the dried powders were then calcined in air at 1150 °C for 2 h on recrystallised alumina crucibles. Finally the calcined powders were ground and sieved through a 425 μm sieve. The second stage involved mixing the starting

* Corresponding author. Tel.: +86 21 5241 2005; fax: +86 21 5241 3903.
E-mail address: rainbow@mail.sic.ac.cn (R.-H. Liang).

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powder with MgO and La_2O_3 in ethanol for 24 h and the dried powders were then calcined in air at 1250 °C for 2 h in air. The calcined powders were again wet ground for 48 h. The dry calcined powders were mixed with polyvinyl alcohol (7 wt.%). The powders were then pressed into pellets 13 mm in diameter and approximately 2 mm (for low frequency measurement) or 5 mm (for microwave frequency measurement) in thickness and sintered between 1350 and 1450 °C for 2 h, depending on the amount of La_2O_3. After sintering, the 2 mm disc samples were polished to 0.5 mm and electroded with silver paint for low frequency electrical measurements.

The phase of these specimens was examined by X-ray diffraction with Cu Kα radiation (Rigaku RAX-10, Japan). The dielectric constant of each sample was calculated from the measured capacitance and the specimen geometry. The capacitance and dielectric loss tangent at low frequency were measured with an inductance–capacitance–resistance (LCR) meter (Agilent 4294A). The dielectric properties at microwave frequency were measured with Network Analyzer (Agilent E8363A). Electron probe microanalyzer (EPMA, JXA-8100A) was used to observe the surface. For the tunable properties, a 1000 V DC power supply was connected to the solartron 1260 frequency response analyzer as the external DC bias electric field up to 1900 V mm^{-1}.

3. Results and discussion

X-ray diffraction was utilized to assess the samples crystallinity and to determine whether or not the samples possessed a solid solution or multiphased structure. Fig. 1 displays the X-ray diffraction patterns of the La_2O_3 doped (0–3.0 wt.%) of $Ba_{0.55}Sr_{0.45}TiO_3$/MgO composites. The result shows these samples possess diphase structure of $Ba_{0.55}Sr_{0.45}TiO_3$ and MgO and there is no La_2O_3 phase formation. The full-width-at-half-maximum (FWHM) of the most intense diffraction peaks increased with increasing La_2O_3 doping content, especially when the doping content is up to 3.0 wt.% La_2O_3. This peak broadening is indicative of a decrease in grain size, which is also can be seen from the EPMA result below.

Fig. 2 shows the surface EPMA micrographs of La_2O_3 doped (0–3.0 wt.%) of $Ba_{0.55}Sr_{0.45}TiO_3$/MgO composite, EDS analysis confirmed that the dark phase contained primarily Mg and O and the light phase contained primarily Ba, Sr, Ti and O which is shown in Fig. 3. And in $Ba_{0.55}Sr_{0.45}TiO_3$ grain, the EDS analysis also detected La peaks and it may indicate the La^{3+} occupation of Ba/Sr site, so there is no La_2O_3 phase formation which is consistent with the result of Fig. 1. From the figure, the grain size of $Ba_{0.55}Sr_{0.45}TiO_3$ and MgO both decrease with the increasing doping content of La_2O_3, especially the doping content is up to 3.0 wt.% and this analysis supported the result of X-ray diffraction. The undoped samples show very rough microstructure and the grain sizes of different site differ greatly. The addition of La_2O_3 made the grain size distribution to be regular.

Fig. 4 shows the dielectric properties of these samples as a function of temperature at low frequency 10 kHz. Table 1 shows the dielectric properties of the samples at low frequency. The dielectric constant decreases with increasing La_2O_3 doping concentration and this is attributed to the La^{3+} occupation of Ba/Sr site which induces a decrease of the curie temperature. The temperature coefficient of capacitance

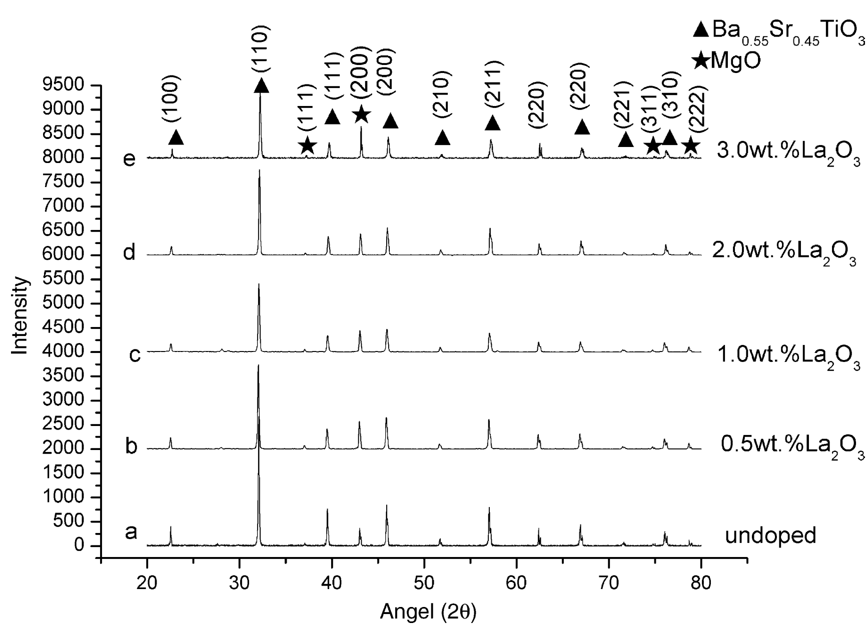

Fig. 1. XRD patterns for all the samples: (a) $Ba_{0.55}Sr_{0.45}TiO_3$/MgO; (b) 0.5 wt.% La_2O_3 doped; (c) 1.0 wt.% La_2O_3 doped; (d) 2.0 wt.% La_2O_3 doped; (e) 3.0 wt.% La_2O_3 doped.