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Microwave dielectric properties of tunable $Ba_{0.5}Sr_{0.5}TiO_3$ and scheelite $AMoO_4$ (A = Ba, Sr) composite ceramics

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ABSTRACT

$(1-x)Ba_{0.5}Sr_{0.5}TiO_3$–$xAMoO_4$ (A = Ba, Sr) ceramics with x = 5, 10, 20 and 30 wt% were prepared by traditional solid-state reaction method. Two crystalline phases, a cubic perovskite structure $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) and a tetragonal scheelite structure $AMoO_4$ were observed by XRD analysis. The microwave dielectric properties of $Ba_{0.5}Sr_{0.5}TiO_3$–$AMoO_4$ composite ceramics were investigated systematically. With increasing content of $AMoO_4$ from 5% to 30%, the dielectric anomalous peaks of ferroelectric–paraelectric phase transition for the composite ceramics are suppressed and broadened. The results show that the composite ceramics exhibited promising microwave properties. Especially, in the $(1-x)Ba_{0.5}Sr_{0.5}TiO_3$–$xSrMoO_4$ system the dielectric constant can be adjusted in the range from 1200 to 132, while maintaining tunability from 38.3% to 6.4% (60 kV/cm) and Q values more than 259 in the gigahertz frequency region. So $(1-x)Ba_{0.5}Sr_{0.5}TiO_3$–$xSrMoO_4$ composite ceramic is a promising candidate for electrically tunable microwave device applications.

1. Introduction

Due to Barium strontium titanate ($Ba_{1-x}Sr_xTiO_3$ or BST) material's nonlinear dielectric response to applied electric fields, It has attracted significant interest as active elements of tunable microwave devices for wireless telecommunications [1,2]. The strong dependence of the dielectric properties on the electric field can be employed to develop tunable microwave devices such as filters, varactors, delay lines, and phase shifters. For applications, materials should exhibit high dielectric tunability ($T = \Delta\varepsilon/\varepsilon > 10\%$) with applied field and low dielectric loss ($\tan\delta < 0.005$) at microwave frequencies [3–5]. But the critical material parameters for many microwave designs are low dielectric constant for impedance matching, low-dielectric loss tangent, and high-dielectric tunability in a certain electric-field range. The high-dielectric loss and high dielectric constant of BST have restricted its application. To meet these requirements, some groups [6–9] have added non-ferroelectric materials (such as MgO, Mg_2SiO_4, and $BaTi_4O_9$) to synthesize BST composites to dilute the dielectric constant meanwhile maintain acceptable dielectric tunability. The experiments of BST-based composites with Mg_2SiO_4 and MgO have been reported to obtain low dielectric constant, but the tunability was reduced to less than 10%. $Ba_{1-x}Sr_xTiO_3$–$BaTi_4O_9$ based composite ceramics had also problems for microwave applications, due to their relative high loss tangent at microwave frequencies. The purpose of this study is to find a new composite BST system which has high tunability, low dielectric constant and high Q value at microwave frequencies.

In this study, based on $Ba_{0.5}Sr_{0.5}TiO_3$ (BST50) with addition of $AMoO_4$ solid solution was investigated. $AMoO_4$ was selected because of the $AMoO_4$ samples exhibited dielectric constant of 7–11, quality factor of 37,000–90,000 GHz [10]. In the previous work [11], we have investigated the dielectric properties of $Ba_{0.4}Sr_{0.6}TiO_3$–$BaMoO_4$ composite ceramic, and better microwave properties were obtained. However, the tunability is not high enough. In the present work, based on $Ba_{0.5}Sr_{0.5}TiO_3$–$AMoO_4$ (A = Ba or Sr) composite system, the dielectric constant, loss, and tunability of composites as a function of volume fractions of dielectric material are investigated systematically, and in comparison with the different effect of $BaMoO_4$ and $SrMoO_4$ on properties of $Ba_{0.5}Sr_{0.5}TiO_3$ composite ceramics. So we tried to systematically investigated to evaluate the general trends of dielectric properties of BST50–$AMoO_4$ composites and then optimized good dielectric behavior to satisfy the requirements of tunable devices.

2. Experimental processes

The ceramic samples were synthesized using the conventional solid-state reaction. High-purity $BaTiO_3$ (99.9%), $SrTiO_3$ (99.9%), $BaCO_3$ (99.9%), $SrCO_3$(99.9%) and MoO_3 (99.99%) were used as starting materials for the synthesis of $Ba_{0.5}Sr_{0.5}TiO_3$ (BST50) powders at 1100 °C and $AMoO_4$ (A = Ba and Sr) powders at 700 °C, respectively. After that, the powders were weighed according to stoichiometric ra-

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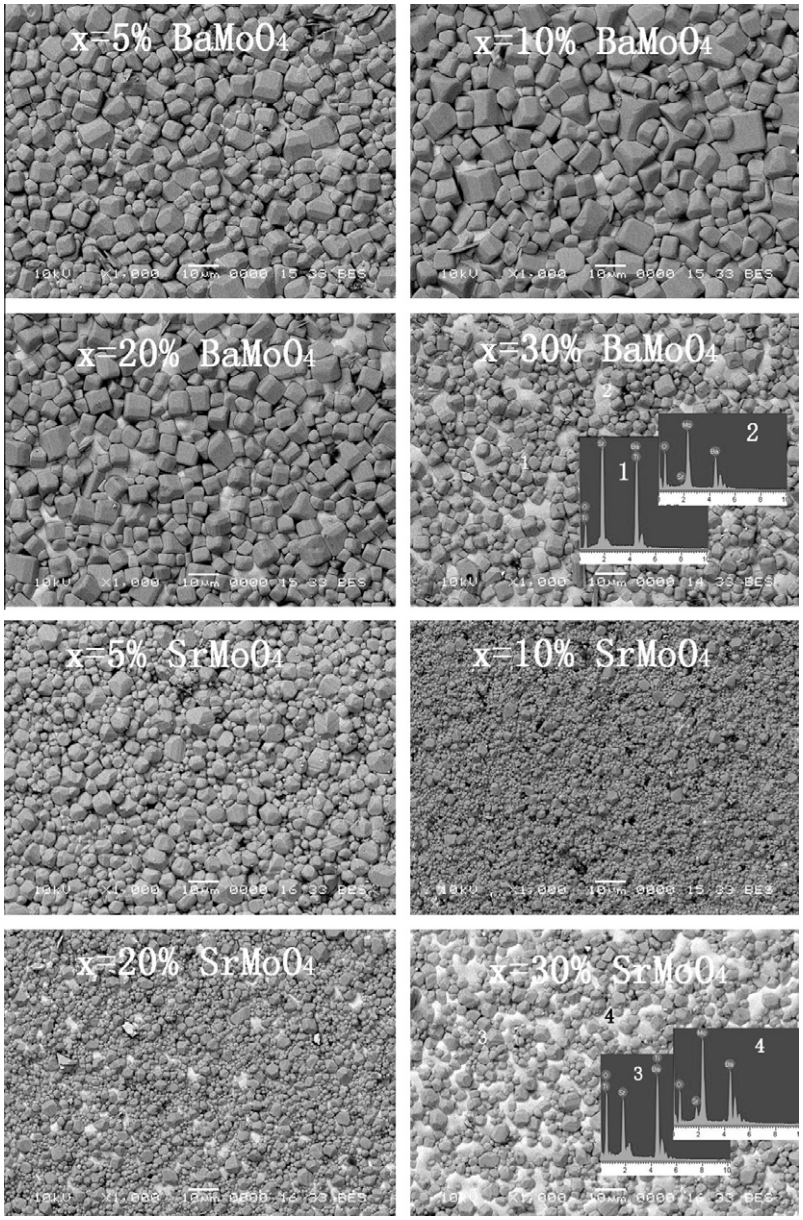

Fig. 1. Backscattered images of $(1-x)Ba_{0.5}Sr_{0.5}TiO_3$–$xAMoO_4$ composite ceramics samples.

tio $(1-x)Ba_{0.5}Sr_{0.5}TiO_3$–$xAMoO_4$ (where x = 0, 5, 10, 20, 30 wt%) and mixed in polypropylene bottles with zirconia grinding media for 24 h. Ethanol was added until a thick slurry was obtained. The mixture was then ball-milled in a planetary machine and dried at 110 °C for 12 h. The dried powders, mixed with 8 wt% polyvinyl alcohol (PVA), were pressed into pellets. Samples for low frequency dielectric measurement are 10 mm in diameter and 1.2 mm in thickness, while those for microwave frequency measurements have dimensions of 15/7 mm in diameter/thickness, respectively. The green pellets were burned out at 550 °C for 6 h in air to remove the solvent as well as the binder, and then sintered at 1350 °C for 4 h in air.

X-ray diffraction (XRD) (D8 Advanced, Bruker, Germany) was used to identify the phase structure. Scanning electron microscopy (JSM EMP-800, JEOL, Tokyo, Japan) was used to characterize the microstructure. The samples were polished to 1 and 0.15 thicknesses and gold electrodes were sputtered on both sides of the samples for dielectric property measurements. Temperature dependences of dielectric constant and loss at 10 kHz were measured in the temperature range from −170 °C to 140 °C using an E4980A LCR meter (Agilent, Palo Alto, CA). The dielectric constant as a function of electric field was tested using a Keithley 2410 (Cleveland, OH) high-voltage source and a TH2816A LCR (Tonghui Electronics, Changzhou, China) analyzer. Dielectric properties at microwave frequencies were obtained using the Hakki–Coleman dielectric resonator method with a network analyzer (HP8753E, Agilent) combining a resonating cavity. The values of ε and Q were calculated from the resonant frequency and the geometric dimensions of the samples.

3. Results and discussions

The micro structural evolution of sintered samples $(1-x)Ba_{0.5}Sr_{0.5}TiO_3$–$xAMoO_4$ at 1350 °C is presented in Fig. 1. From the backscattered electron images, two phases can be observed in these samples and all exhibit a quite dense microstructure. The gray phase consists of primarily Ba, Sr, Ti and O, while the white phase consists of primarily Ba, Sr, Mo, O. So the gray grains are BST phase, and the homologous white grains are AMoO4 phase. This EDS result in dictated that inter-diffusion between BST and AMoO4 occurred during the sintering process. All samples have dense and homogeneous microstructures. Obviously, the connectivity levels of the BST phase decreased gradually with increasing of $AMoO_4$ content. This is the major factor in the decrease of its tunability [12].

Fig. 2 shows the XRD patterns of $(1-x)Ba_{0.5}Sr_{0.5}TiO_3$–$xAMoO_4$ (A = Ba or Sr; x = 0.05, 0.1, 0.2 and 0.3) samples sintered at 1350 °C/4 h. The cubic perovskite BST phase and scheelite $AMoO_4$ phase were clearly observed for all compositions. Both $BaMoO_4$