Available online at www.sciencedirect.com

SciVerse ScienceDirect

Ceramics International 39 (2013) S335–S339

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Dielectric properties of $Ba_{1-x}Sr_xTiO_3$ ceramics prepared by microwave sintering

Yang Yu, Xusheng Wang*, Xi Yao

Functional Materials Research Laboratory, Tongji University, Shanghai 200092, China

Available online 27 October 2012

Abstract

A comparative study on the dielectric properties of $Ba_{1-x}Sr_xTiO_3$ ($x=0.1$–0.6) ceramics prepared by microwave sintering (MS) and conventional sintering (CS) has been done. It was found that MS samples need lower temperature and much shorter time than CS samples to obtain the same degree of densification. Compared with CS samples, MS samples possessed smaller grain size, better densification and more uniform grain growth. The dielectric properties of the samples were measured as a function of temperature. It was observed that the dielectric constant was higher for MS samples compared with that of CS samples especially in the ferroelectric phase.

Keywords: A. Microwave processing; C. Dielectric properties; Barium strontium titanate

1. Introduction

Barium strontium titanate (BST), $Ba_{1-x}Sr_xTiO_3$ is a perovskite-structured ferroelectric material, and it is a solid solution composed of barium titanate and strontium titanate. For its favorable dielectric and ferroelectric properties such as high dielectric constant, alterable Curie temperature, low dielectric loss and high tunability of dielectric behavior, it has been widely used in preparation of dynamic random access memories (DRAM), dielectric capacitors, microwave phase shifters, transducers, positive temperature coefficient resistors (PTC) [1–4].

Many efforts have been put to improve the microstructure and electrical properties of BST ceramics synthesized by conventional and other chemical processes [5–7]. However, very limited literature reported is available on the microwave sintering of BST ceramics, especially for a wide range of Ba/Sr ratio [8–11].

Microwave sintering is a unique technique alternative to the conventional sintering. This technique requires less time and lower temperature to achieve the same quality with ceramics as sintered by conventional route. Furthermore smaller grain sizes and more uniform microstructure can be developed by microwave sintering [8], by which the heat is generated internally within the material through microwave–material interaction instead of originating from external sources [9]. Microwave sintering heated rapidly as the material is heated by energy conversion rather than through energy transfer, and it is very uniform as the microwave–material interaction occurred from the inside to the outside simultaneously rather than energy transfer from the outside to the inside.

Here we report our results on microstructure and dielectric properties of $Ba_{1-x}Sr_xTiO_3$ ($x=0.1$–0.6) ceramics sintered by microwave and conventional methods.

2. Experimental

$Ba_{1-x}Sr_xTiO_3$ ($x=0.1$–0.6, denoted as BST10, BST20 for $x=0.1$, 0.2…) powders were synthesized by the conventional solid-state reaction of barium titanate ($BaTiO_3$, 99.9%) and strontium titanate ($SrTiO_3$, 99.9%) powders. Stoichiometric mixtures of the raw powders were mixed in ethanol medium with zirconia balls and ball-milled for 24 h. After drying, the mixtures were calcined at temperatures ranged 1100 °C to 1200 °C for 2 h, and remilled for 24 h to reduce the particle size for sintering, and then granulated with polyvinyl alcohol (PVA) which is used as a

*Corresponding author. Tel.: +86 21 65980544; fax: +86 21 65985179.
E-mail address: xs-wang@tongji.edu.cn (X. Wang).

0272-8842/$ - see front matter © 2012 Elsevier Ltd and Techna Group S.r.l. All rights reserved.
http://dx.doi.org/10.1016/j.ceramint.2012.10.089

Table 1
Detailed sintering temperature.

Component	Microwave sintering (°C)	Conventional sintering (°C)
BST60	1300	1320
BST50	1280	1300
BST40	1280	1300
BST30	1260	1280
BST20	1260	1280
BST10	1220	1240

binder. The granulated powders were pressed into discs in diameter of 10 mm and thickness of 1.0 mm.

Jae-Ho Jeon has reported the effect of sintering temperature on microstructure and dielectric constant of $Ba_{1-x}Sr_xTiO_3$ [12]. In this paper, we select a suitable temperature for each component and each sintering method instead of a same temperature. For conventional sintering, one set of these pellets were sintered at temperatures ranged 1240 °C to 1320 °C for 4 h, maintaining 3 °C/min heating rate. Another set of pellets were sintered by a microwave furnace (2.45 GHz, 1.4 kW) at temperatures ranged 1220 °C to 1300 °C for only 0.5 h of soaking time. The heating rate was maintained at 30 °C/min (by controlling the input power). The detailed sintering temperature is listed in Table 1.

For electrical characterization, the ceramic pellets were polished, coated with silver paste, and fired at 600 °C for 10 min.

Their microstructure morphology was obtained by scanning electron microscopy (SEM, JSM EMP-800, JEOL, Japan), and phase structure was analyzed by an X-ray diffraction (XRD, D8 Advance, Bruker, Germany) with CuKα radiation. The temperature-dependence of dielectric properties was investigated with an LCR meter (HP4284A, Agilent, Palo Alto, CA). The polarization–electric field hysteresis loops were measured with a ferroelectric analyzer (Premier II, Radiant, USA).

3. Results and discussion

The XRD patterns of the conventional and microwave synthesized samples are shown in Fig. 1. This confirms the formation of a single phase perovskite BST structure. As Sr content increases, the diffraction peaks move toward high-angle direction, and the phase structure undergoes a transition from tetragonal (BST10, BST20, BST30) to cubic (BST40, BST50, BST60) gradually. MS samples achieve the same intensity of peak (110) as CS samples, which indicates that they have the same crystallization.

The surface microstructure of the MS and CS samples are shown in Fig. 2 and the grain sizes are listed in Table 2. Compared with CS samples, MS samples show smaller grain size, better densification and more uniform grain growth as shown in SEM images. The rapidity and

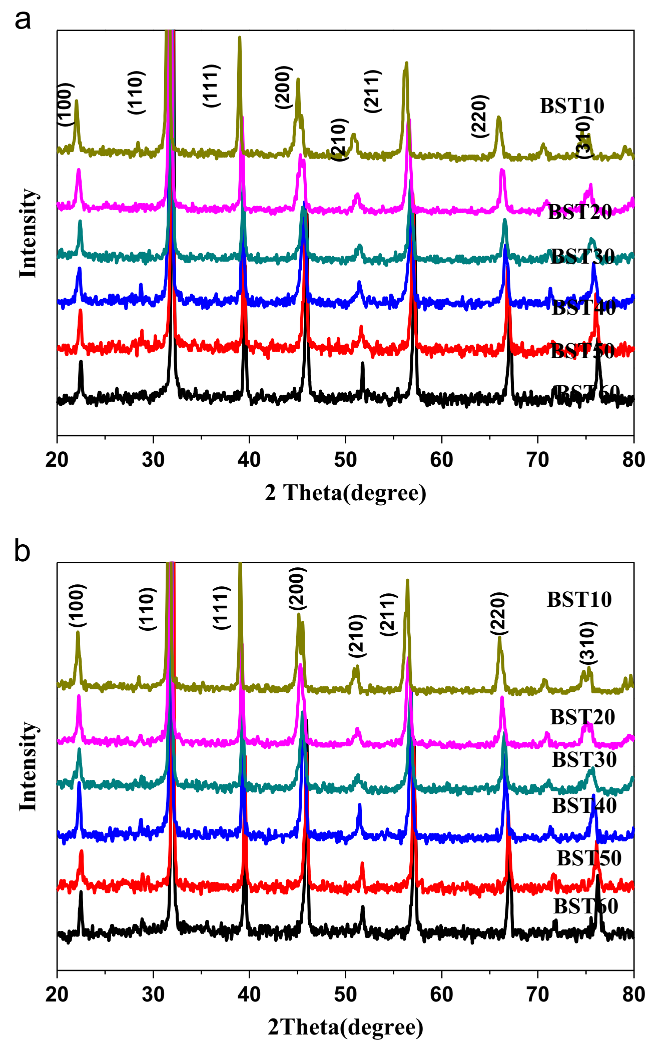

Fig. 1. XRD patterns of BST ceramics: (a) MS samples and (b) CS samples.

uniformity of microwave sintering avoids undesirable grain growth and provides a finer and more uniform microstructure.

Temperature dependence of relative dielectric constant and dielectric loss of MS and CS samples are shown in Fig. 3, and dielectric parameters are summarized in Table 3. Compared with the CS samples, MS samples possess a higher maximum dielectric constant ε_m for they have smaller grain size, which agrees with those reported elsewhere [11]. For BST10, BST20, BST30 and BST40, the Curie temperature of samples sintered by the microwave method was higher than those sintered by the conventional method, and the effect was opposite for BST50 and BST60. Compared with the CS samples, the dielectric constant (25 °C, 10 kHz) of MS samples increased about 20%, 35%, 25%, 9.2%, 2.0% and 1.5% for BST10, BST20, BST30, BST40, BST50 and BST60, respectively. BST ceramics were in the ferroelectric phase at room temperature (25 °C) when $x \leq 0.35$ [13], it was observed that the dielectric constant was higher for MS samples