STRUCTURAL AND DIELECTRIC PROPERTIES OF BARIUM STRONTIUM TITANATE ($Ba_{(1-X)}Sr_XTiO_3$) CERAMICS

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ABSTRACT

Structural and dielectric properties of barium strontium titanate ($Ba_{(1-x)}Sr_xTiO_3$) ceramics with x= 0.25, 0.5 and 0.75 were investigated. The $Ba_{(1-x)}Sr_xTiO_3$ ceramics were synthesised by solid state reaction method. Microstructure, surface morphology and dielectric properties of the synthesised ceramics were examined using XRD, SEM and Impedance Spectroscopy respectively. XRD results revealed that all samples contained $BaTiO_3$ as primary phase with tetragonal perovskite crystal structure. The crystallite size slightly increased with the increasing of Sr contents. SEM micrographs showed that the microstructure become denser when Sr contents increased. Impedance spectroscopy showed dielectric constant for all samples were decreased with increasing temperature up to 200°C.

ABSTRAK

sifat-sifat struktur dan dielektrik barium strontium titanate (Ba (1-x) Sr_xTiO_3) seramik dengan x = 0.25, 0.5 dan 0.75 telah disiasat. The Ba (1-x) seramik Sr_xTiO_3 telah disintesis melalui kaedah tindak balas keadaan pepejal. Mikrostruktur, permukaan morfologi dan dielektrik sifat-sifat seramik disintesis telah diperiksa menggunakan masing-masing XRD, SEM dan Impedance Spektroskopi. keputusan XRD mendedahkan bahawa semua sampel yang terkandung $BaTiO_3$ sebagai fasa utama dengan tetragonal struktur perovskit kristal. Saiz crystallite sedikit meningkat dengan peningkatan kandungan Sr. SEM mikrograf menunjukkan mikrostruktur menjadi lebih padat apabila isi kandungan Sr meningkat. Galangan spektroskopi menunjukkan pemalar dielektrik untuk semua sampel telah menurun dengan peningkatan suhu sehingga 200°C.

Keywords: $Ba_{(1-x)}Sr_x$ TiO_3, different Sr contents, structural, surface morphology, dielectric constant.

INTRODUCTION

In recent years, Barium Titanate ($BaTiO_3$) ceramics have been known as the important electronic materials for use in capacitors, thermistor and piezoelectric transducers [1-3]. These are due to their ferroelectric properties. Nowadays, in order to improve these properties, $BaTiO_3$ ceramics have been partial substituted with other elements [4-7].

Partial substitution of Ba by strontium (Sr) results in high dielectric constant, nonlinear variation of dielectric constant with electric field, ferroelectricity and pyroelectricity properties [8-10]. Therefore barium strontium

titanate ($Ba_{(1-x)}Sr_xTiO_3$) ceramic is a potential candidate for dynamic random access memories (DRAM), Non-Volatile Ferroelectric Random Access Memories (NVFRAM) microwave phase shifters, tunable filter, multilayer and voltage tunable capacitor, oscillators and uncooled infrared sensors [11-13].

($Ba_{(1-x)}Sr_xTiO_3$) is a ferroelectric materials with a Curie temperature decreases almost linearly with increasing x. $Ba_{(1-x)}Sr_xTiO_3$ showed wide range of dielectric constant varying from a few hundred to thousands depending on Ba/Sr ratio, grain size and temperature [14]. Incorporating rare earth oxides (Sr) into the perovskite structure is an effective way to improve dielectric properties of barium strontium titanate ceramics. In order to attain the desired electrical characteristics, high chemical purity and uniform microstructure are the most important features for $Ba_{(1-x)}Sr_xTiO_3$ ceramics [15].

In this present work, we report the effects of Sr contents on the structural, surface morphology and dielectric properties of the $Ba_{(1-x)}Sr_xTiO_3$ ceramics prepared with solid state reaction method.

EXPERIMENTAL METHODS

Sample preparation

The chemical compositions of the samples are given by the formula ($Ba_{(1-x)}Sr_xTiO_3$) where x= 0.25, 0.5 and 0.75. High purity of Barium Carbonate ($BaCO_3$, Sigma-Aldrich 99.9%), Strontium Oxide (SrO, Sigma-Aldrich 99.9%) and Titanium Oxide (TiO_2, Sigma-Aldrich 99.9%) powders were used as starting materials. These powders were weighed out in stoichiometric proportions and ball-milled at 300 rpm for 12 hours. Ethanol was used as a milling medium. Then the milled samples were dried at 100°C for 24 hours and were calcined at 1200°C for 6 hours in a high purity alumina crucible. The calcined samples were crushed into fine powders using a mortar. Then they were pressed into a pellet with 10 mm diameter and 2.2 mm thickness. The green pellets were sintered at 1200°C for 2 hours using an alumina boat/plate. For electrode formation, silver paste was applied on both surfaces of the samples.

CHARACTERIZATIONS

The microstructure of the calcined powders was determined by X-Ray Diffraction (XRD) technique using a PANalytical diffractometer (CuK_α radiation) with a 2θ range of 10 - 80° using 0.02° steps and a 0.8 s count time per step with operating conditions of 40 kV and 40 mA. Surface morphology of all samples was conduction on a Philips XL-30 Scanning Electron Microscopy (SEM) respectively. The dielectric properties of the pellet samples were measured with High Performance Frequency Impedance Analyzer (Novovontrol). The crystallite sizes were refined from the diffraction data using Rietveld refinement in Highscore Plus (PANalytical, Netherland). In this calculation, the broadening from emission and instrumental parameters have been considered and subsequently corrected.

RESULTS AND DISCUSSION

Figure 1 shows the XRD diffractogram of $Ba_{(1-x)}Sr_xTiO_3$ powders with different strontium contents (sample BST 0.25, sample BST 0.5 and sample BST 0.75) respectively. Phase identification analysis confirmed that all samples were related to $BaTiO_3$ phase (JCPD: 98-004-6166) with tetragonal perovskite crystal structure. No other secondary phase was detected, which suggest that the solid state reaction was completed. This result is

consistence with the previous study of the synthesis of $BaTiO_3$ using the solid state method [16]. The peaks at 32° shifted to the right when Sr contents were increased from 0.25 to 0.75.

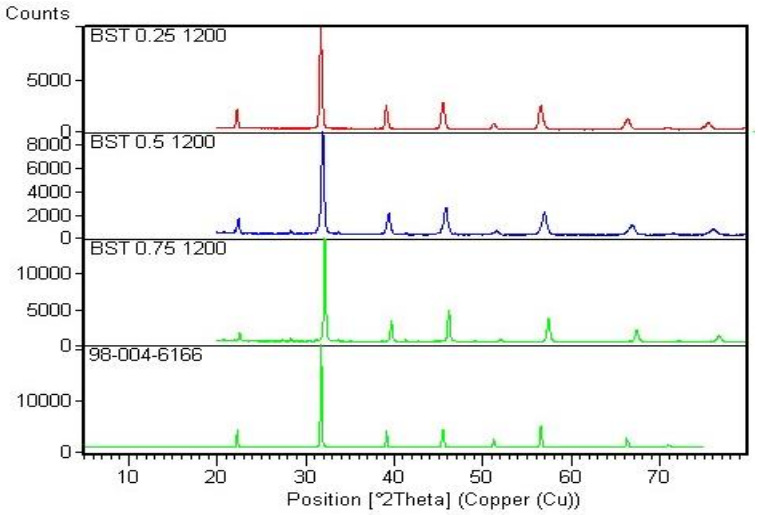
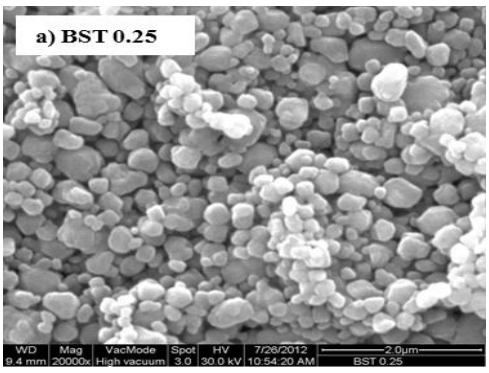
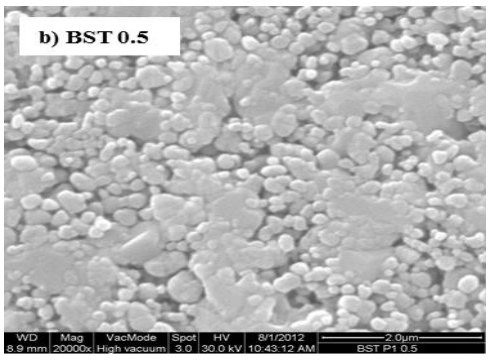


FIGURE 1. XRD diffractograms of synthesized $Ba_{(1-x)}Sr_xTiO_3$ powders with Sr contents of x = 0.25, 0.5 and 0.

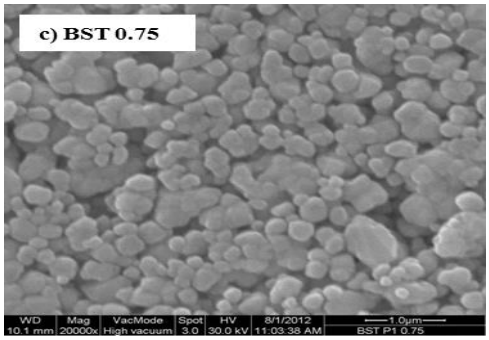
The surface morphologies of $Ba_{(1-x)}Sr_xTiO_3$ ceramics with various Sr contents are shown in Figure 2. Sample a (BST 0.25) and b (BST 0.5) exhibits a dense microstructure and uniform grain size. However sample c (BST 0.75) were denser and its grains were joined together with no grain boundary were seen. The average grain size increased as the content of Sr is increased. The grain size distribution was narrowed down with increasing of Sr contents. The substitution of Sr2+ ions for the perovskite structure causes the lattice deformation. The lattice deformation restrains the grain growth of $Ba_{(1-x)}Sr_xTiO_3$ ceramics and thus brings the fine grain structure [6].



(a)



(b)



(c)

FIGURE 2. SEM micrographs of (a) BST 0.25, (b) BST 0.5 and (c) BST 0.75

sintered at 1200°C.

Figure 3 shows the dielectric constant of $Ba_{(1-x)}Sr_xTiO_3$ (x= 0.25, 0.5 and 0.75) measured at high frequency (1Mhz) and different temperatures (25°C – 200°C). It can be noted that the dielectric constant at room temperature (25°C) enhances at first and decreased with increasing temperature. Dielectric constant of BST 0.25 at room temperature was 71.0 and decreased to 19.8 when temperature rises to 200°C. BST 0.5 shows that dielectric constant at room temperature is 84.1 and decreased to 63.1 at 200°C. Sample BST 0.75 exhibit high dielectric constants (88.6) at room temperature and quickly decreased when temperature increased up to 200°C (55.7). All samples revealed that dielectric constant were decreased with increasing of temperature.

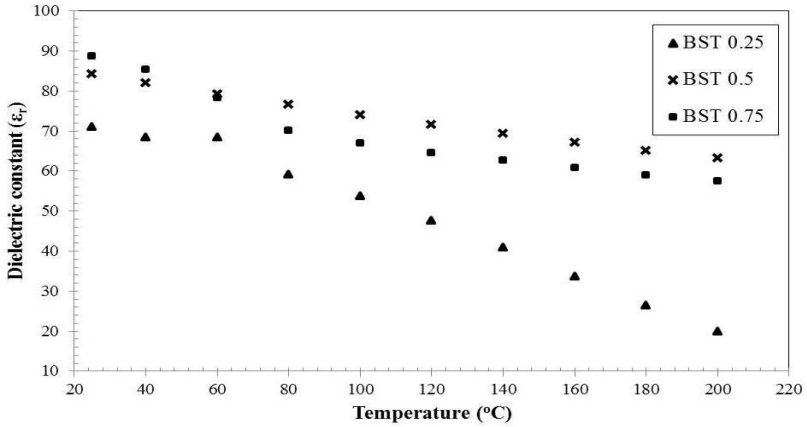


FIGURE 3. Dielectric constant of BST with x= 0.25, 0.5 and 0.75 at different temperatures.

CONCLUSIONS

$Ba_{(1-x)}Sr_xTiO_3$ ferroelectric ceramics were successfully prepared from powders synthesized using solid state reaction method with no secondary phase were detected. The different Sr contents affected the microstructure and morphology of $Ba_{(1-x)}Sr_xTiO_3$ ceramics. According to Chen et al. 2012, the grains growth slowly and joined together (agglomerate) when the temperature was increased to 200°C. It caused the lattice deformation of $Ba_{(1-x)}Sr_xTiO_3$ structure. The different contents of Sr affected the dielectric constants of all the BST samples. Dielectric constants for BST 0.25 decreased from 71.0 to 19.8, BST 0.5 decreased from 84.1 to 63.1 and BST 0.75 decreased from 88.6 to 55.7. Dielectric constants of BST 0.25 were dropped quickly and it can be claimed that BST 0.25 more sensitive than BST 0.5 and BST 0.75.

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