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Original Research Paper

Effect of pressure of pelletization on dielectric properties of Bismuth Titanate prepared by sol-gel synthesis

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ABSTRACT

Ferroelectric Bismuth Titanate ($Bi_4Ti_3O_{12}$) was prepared by sol-gel synthesis method. Bismuth nitrate and titanium isopropoxide were used as the starting precursors in isopropanol. The annealing of precipitate powders was done at 400 °C for 4 h. Effect of pressure of pelletization on the dielectric properties of $Bi_4Ti_3O_{12}$ was studied. The pressure was varied from 585 MPa to 1365 MPa during pellet preparation. The orthorhombic structure of $Bi_4Ti_3O_{12}$ was obtained after sintering at 800 °C for 4 h, which was confirmed by X-ray diffraction. Dielectric studies have been done in a wide frequency (20 Hz–20 MHz) and temperature (100–400 °C) range.

1. Introduction

Bismuth Titanate (BTO) is a material with wide applications in non-volatile ferroelectric random access memory devices (FRAM) and WiMAX/WLAN Antenna [1,2]. It is member of BLSFs (Bismuth Layered-Structure of Ferroelectrics) with a high curie temperature (675 °C). It shows spontaneous polarization value of 50 μC/cm^2, and coercive field of 50 kV/cm [3,4], which gives it a potential applicability as a suitable candidate for high-temperature piezoelectric device.

BTO belongs to the layered perovskite compound which is the Aurivillius [5–7] family, used as alternative lead – free ferroelectric and piezoelectric material with the general formula $(Bi_2O_2)[A_{m-1}(B)_mO_{3m+1}]$, which consists of $(Bi_2O_2)^{2+}$ sheets alternating with $(Bi_2Ti_3O_{10})^{2-}$ perovskite-like layers. In general formula, *m* is the number of octahedral stacked between the direction perpendicular to the sheets, and A and B are the 12- and 6-fold coordination sites of perovskite slab, respectively [5,6].

BTO has been prepared by Chemical Precipitation [8–10], hydrothermal [11–14], Solid state [15–17], flux method [18,19], conventional mixed oxide method [20], oxalate method [21], self-propagating high-temperature synthesis [22], combustion route [23], oxidant peroxo method [24], and sol – gel synthesis [25–27]. Sol – gel synthesis is a low temperature synthesis method, it is inexpensive and gives good homogeneity of composition. Even though BTO ceramics are studied widely, the effect of pressure of pelletization on its properties is not reported. For these reasons the study focuses on the effect of compaction pressure on the dielectric properties of BTO.

The present work is aimed at the study of effect of pressure of pelletization on the Bismuth Titanate ceramic prepared by sol-gel method. The phase formation has been confirmed by X-ray diffraction (XRD). The morphology and elemental composition have been studied by scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDXS) respectively. The dielectric constant (ε_r), loss tangent (tan δ), DC resistance (R_{DC}) and capacitance (C) have been measured as function of temperature (100–400 °C) over a wide range of frequency (20 Hz–2 MHz) using a high resolution dielectric analyzer (Novocontrol Impedance analyzer) and sample holder with silver electrodes. The density of samples was measured using Archimedes principle with toluene as an immersion medium.

2. Experimental

BTO ceramics were synthesized by the sol-gel method. The schematic diagram for the synthesis is shown in Fig. 1 the starting precursors used include Bismuth (III) Nitrate Pentahydrate (Bi $(NO_3)_3$ $5H_2O$) (Merck), Titanium (IV) Isopropoxide (Ti $(OC_3H_7)_4$ (Sigma – Aldrich) and 2-propanol (Merck). The starting materials Bismuth nitrate and Titanium isopropoxide were mixed in 4:3 proportion and dissolved in 2-propanol under vigorous stirring for 4 h to obtain a sol, then stirring was stopped in a

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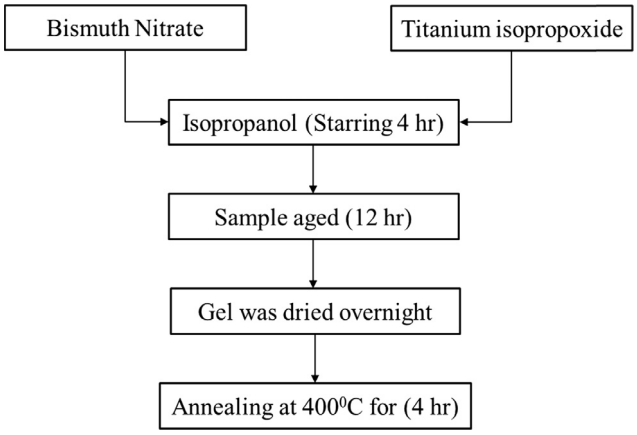

Fig. 1. Schematic diagram for the synthesis of BTO powders by sol-gel method.

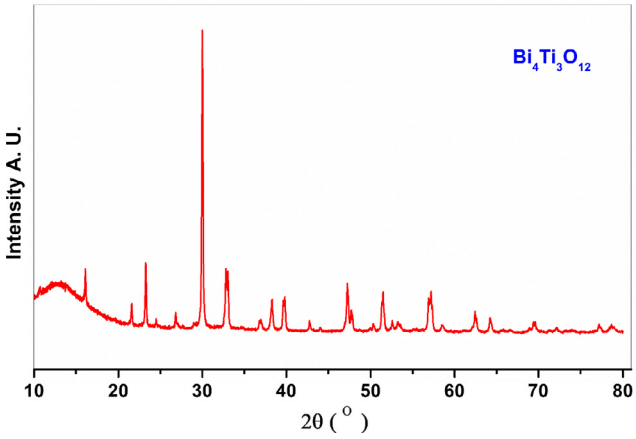

Fig. 2. XRD pattern of the powder of BTO thermally treated at 800 °C.

closed system. The samples were aged for 12 h, after that an excess of water was added to hydrolyse completely the system. The resulting gel was dried overnight. After that annealing was done at 400 °C for 4 h. The powder of the material was cold pressed into circular disc-shaped pellets of using hydraulic press (with PVA as binder) at the pressure of 585 MPa, 975 MPa and 1365 MPa, then sintered at 800 °C for 4 h. followed by natural cooling of the furnace. The PVA burnt out during high-temperature sintering. Table 1 summarizes the reported values of annealing temperature and sintering temperature for BTO samples with different synthesis method. From the table it is clear that the sol – gel synthesis methods requires lower annealing and sintering temperatures as compared to other methods, which is advantageous. Finally, the sintered pellets of 2 mm thickness and 8 mm diameter were polished with fine emery paper to make the opposite faces flat and parallel. These pellets were coated with high-purity silver paint and dried at 100 °C for 1 h to remove the moisture from the sample prior to electrical measurements. Dielectric measurements were carried out using sample holder with silver electrodes.

3. Results and discussion

3.1. X-ray diffraction

The X-ray diffraction data for BTO were collected using Burker AXS D8 Advance diffractometer. The X-ray source is a 2.2 KW Cu anode. The running condition for the X-ray tube was 40 kV and 40 mA. The wavelength, $\lambda_{K\alpha1}$ = 1.5405 Å, $\lambda_{K\alpha2}$ = 1.4433, Kα1/Kα1 = 0.5, 2θ range between 10° to 80° with step size 0.009°. The lattice parameters were calculated using Rietveld Refinement using FULLPROF programme suit [28]. A pseudo-Voigt function was chosen to generate the line shape of diffraction peaks. Fig. 2 shows the XRD pattern of BTO. The XRD pattern was indexed by comparing with standard JCPDS data (JCPDS No 35-0795). The diffraction pattern consists of a number of sharp and small peaks. The XRD pattern of BTO powder exhibits splitting of the higher angle peaks confirming the existence of orthorhombic phase of the material. The broad XRD peak within the 2θ angle 10–20° observed in XRD pattern may be due to instrumental factor as position – sensitive detector is used and not due to amorphous material. The results of Rietveld refinement are presented in Fig. 3 The crystal structure of BTO was refined by Rietveld fitting to orthorhombic symmetry with space group Aba2 with lattice parameter a = 32.8137 Å,

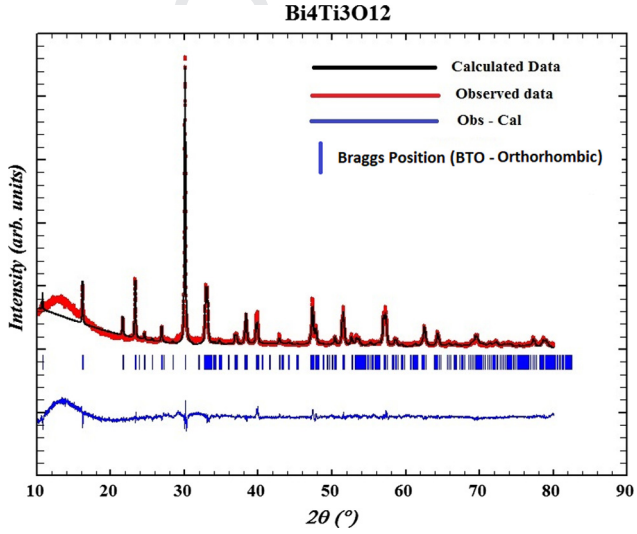

Fig. 3. Rietveld fitted XRD data file of BTO.

Table 1
Annealing temperature and sintering temperature for various BTO samples with different synthesis methods.

Synthesis technique	Annealing temperature (°C)	Time (h)	Sintering temperature (°C)	Time (h)	Dielectric constant (100 KHz) 400 °C	Ref.
Co-precipitation	500–800	1	–	–	–	[8]
Solid-state	700–850	24–48	–	–	–	[16]
	–	–	1000	2	–	[17]
Flux method	600–900	2	–	–	–	[18]
Self-propagating high temperature synthesis	1000	1/2	–	–	200	[22]
Combustion	800	3	1050	2	48	[23]
Sol-gel	400	4	800	4	369	Our paper

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