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Fabrication and characterization of bismuth sodium titanate ceramics by high-energy ball milling technique

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Abstract

$Bi_{0.5}Na_{0.5}TiO_3$ (BNT) ceramics with rhombohedral structure were synthesized by a solid-state mixed oxide technique. The material properties of BNT powders and BNT ceramics are found to depend on the milling time via a high-energy ball milling technique, calcination and sintering processing. The results show that the spherical and rounded shapes of BNT nanopowders are prepared by milling for 30 min and 60 min and calcined at 750 °C for 2 h. The BNT ceramics have a uniform grain size of 4.7 μm after sintering at 1125 °C for 2 h and milling for 60 min, and exhibit a maximum dielectric constant of 489, and a dielectric loss of 0.013. The value of relative density is 95% of theoretical density.

Keywords: A. Sintering; Bismuth sodium titanate; High-energy ball milling; Nanopowders

1. Introduction

Bismuth sodium titanate, $Bi_{0.5}Na_{0.5}TiO_3$ (BNT) is a ferroelectric material, which is interesting because it has a large remanent polarization $P_r = 38\ \mu C/cm^2$, and coercieve field $E_c = 73$ kV/cm at room temperature with a perovskite structures of ABO_3 type in which A-site is occupied by the Bi^{3+} and Na^+ ions, while Ti^{4+} are accommodated at the B-sites [1–5].

BNT ceramics have a rhombohedral phase at room temperature. The BNT phase transforms to an antiferroelectric phase above 220 °C and to a paraelectric phase at 320 °C. The phase above 320 °C is a tetragonal and changes to a cubic phase above 520 °C [6]. BNT ceramics are widely used in sensors, ultrasonic transducers, ferroelectric random access memories and electronic devices [7–9]. BNT powders are prepared by many methods such as hydrothermal processing [10], the citrate method [11], sol–gel techniques [12,13] and the conventional solid-state method [14,15]. All methods have been used to fabricate the ceramics and develop the material properties such as particle size, grain size and porosity, and also improve a lower calcination and sintering temperature [16]. Among all, the conventional solid-state method via a high-energy ball milling technique is a simple and low cost synthesis method. Furthermore, this technique is also successful for the preparation of nanopowders [17,18].

The objective of this study is to prepare BNT nanopowders by a solid-state mixed oxide using the high-energy ball milling technique. In addition, the effect of a milling time on microstructure and dielectric properties of BNT ceramics is studied.

2. Experimental

Raw materials of Bi_2O_3, TiO_2, and Na_2CO_3 were used to prepare BNT, with composition $Bi_{0.5}Na_{0.5}TiO_3$. The BNT mixture was ball milled in ethyl alcohol with tungsten carbide grinding media for 30, 60, 90 and 120 min via a high-energy ballmilling method. After drying, the powders were calcined at 750 °C for 2 h with a heating/cooling rate of 5 °C/min. All powders were pressed into pellets at 100 MPa in a 1 cm diameter steel die by using PVA binder. After binder burn out at 500 °C for 2 h with a heating/cooling rate of 5 °C/min, the

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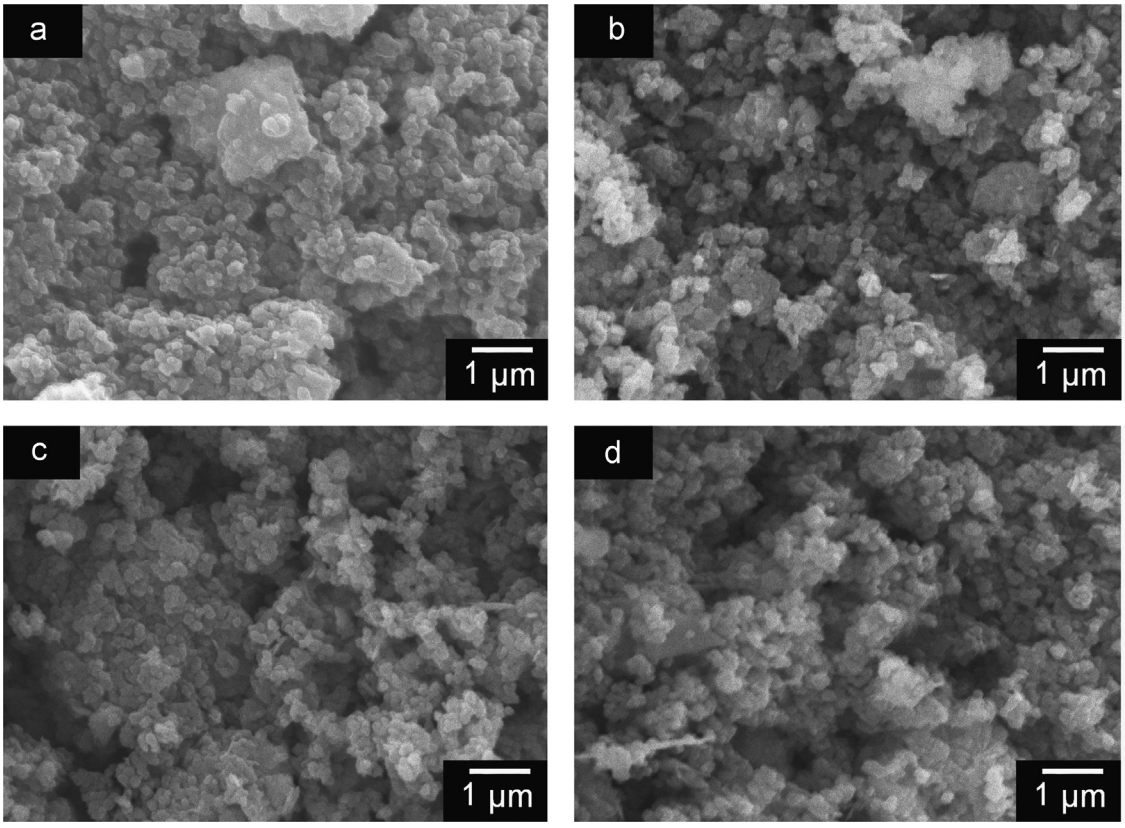

Fig. 1. SEM micrographs of BNT precursor powders via different milling times: (a) 30 min, (b) 60 min, (c) 90 min, and (d) 120 min.

pellets were sintered at 1125 °C for 2 h with a heating/cooling rate of 5 °C/min. Phase identification was performed by X-ray diffraction (XRD) analysis. The microstructure of calcined and sintered samples was observed using a scanning electron microscope (SEM). Density of the sintered samples was measured by using the Archimedes method. The dielectric constant (ε_r) and dielectric loss tangent (tanδ) were measured using an inductance-capacitance-resistance (LCR) meter.

3. Results and discussion

SEM micrographs of BNT precursor powders with milling times for 30–120 min are shown in Fig. 1. The obtained morphology of particles appears to be near spherical and rounded shapes as shown in Fig. 1a–d. The average particle sizes of all BNT precursor powders with various milling times are about 70–200 nm. Furthermore, it was found that the particles seemed to have higher agglomerate with longer milling time.

The XRD analysis shown in Fig. 2 indicates that the single phase with rhombohedral structure appears in all powders. The XRD patterns shows the diffraction peaks around 32.4°, 46.7° and 58.2°, which is also observed by Lee et al. and Razak et al. [19,20], while XRD pattern of BNT precursor powders shows amorphous structure.

SEM micrograph in Fig. 3a shows that BNT nanopowders after calcined at 750 °C for 2 h via milling time for 30 min

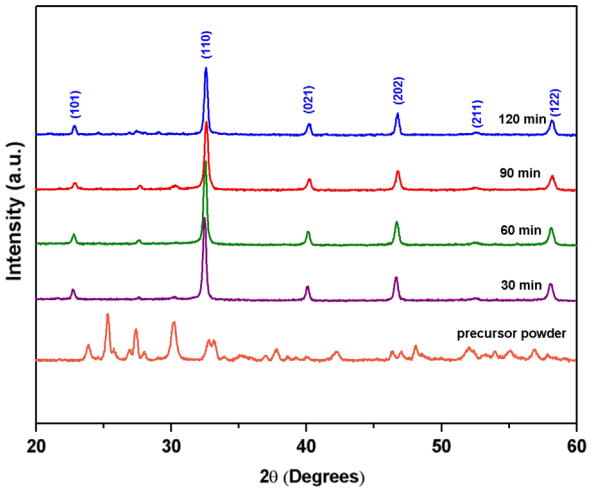

Fig. 2. XRD patterns of BNT precursor powders after milling times for 60 min and BNT powders calcined at 750 °C for 2 h via different milling times.

shows the particle size of approximately 147 nm. The BNT nanopowders via milling time for 60 min are shown in Fig. 3b. The results show that the agglomerated particles were formed, whereas the particle size is about 170 nm. The BNT powders via milling time for 90 min are shown in Fig. 3c. The particle size is in the order of 490 nm, whereas the BNT powders via milling time for 120 min seemed to agglomerate and had a bigger size with the particle size of approximately 717 nm as shown in Fig. 3d.