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Tape casting and electrical characterization of $0.5Ba(Zr_{0.2}Ti_{0.8})O_3$–$0.5(Ba_{0.7}Ca_{0.3})TiO_3$ (BZT–0.5BCT) piezoelectric substrate

S. Abhinay[a], R. Mazumder[a,*], A. Seal[b], A. Sen[b]

[a] Department of Ceramic Engineering, National Institute of Technology, Rourkela, 769008, India
[b] Sensor and Actuator Division, CSIR-Central Glass & Ceramic Research Institute, Kolkata, 700032, India

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ABSTRACT

Recently, piezoelectric wafers/substrates have opened new opportunities for nondestructive evaluation (NDE) of structures in defense, aerospace, and industrial sectors. Such substrates can be easily made by tape casting of slurries where dispersants and binders play a critical role. In the present work, tape casting slurry of BZT-0.5BCT powder has been prepared using MEK-EtOH (solvent), phosphate ester (dispersant), polyvinyl butyral (binder), poly ethylene glycol and benzyl butyl pthalate (plasticizer). It was found that 1 wt.% phosphate ester is sufficient for effective dispersion of BZT-0.5BCT to obtain a stable slurry. Variation of PVB content shows that at least 3 wt.% PVB is required to obtain the workable green tape. The highest density (94% of true density), relative permittivity (1700 at 1 kHz) and the high values of strain (0.23%) have been obtained for tape with 3 wt.% PVB content and 65% solid loading after firing at 1500 °C/4 h.

1. Introduction

Oxide materials with perovskite structure having the general formula ABO_3 form the backbone of the piezoelectric industry. Lead-based materials (PZT, PMN) have dominated in the piezoelectric market because of their excellent dielectric, piezoelectric properties and flexibility regarding property alteration by compositional modifications [1,2]. The amount of lead in the usual lead-containing piezoceramics is more than 60 wt.%. The recent concern about environmental pollution and toxicity of lead has generated the renewed interest in developing lead free ferroelectrics. The breakthrough made by Liu and Ren in $BaTiO_3$-based ceramics with co-dopants of Zr, and Ca has been offered a significant impact on the development of lead-free piezoceramics [3]. The value reported by Liu and Ren [3] for $Ba(Zr_{0.2}Ti_{0.8})O_3$-$0.5(Ba_{0.7}Ca_{0.3})TiO_3$ (BZT-0.5BCT) has been reached more than 500 pCN^{-1}, comparable to that of soft PZT ceramics. Since a long time, piezoelectric materials have been coming into the widespread use in medical ultrasound, underwater sonar systems, smoke detector buzzers [4]. Recently, piezoelectric wafers/substrates have opened new opportunities for nondestructive evaluation (NDE) of structures in defense, aerospace and industrial sectors [5]. There is a requirement for monitoring of structural health and identification of damage for a variety of utilizations running from composite flying vehicles to common and mechanical building frameworks. Structural health monitoring (SHM) or integrated vehicle health monitoring (IVHM) requires small, light-weight, minimally invasive sensors that can be embedded in or mounted on the surface of the structure [6,7]. The applications in SHM often involve piezoelectric ceramics in the form of wafers/substrates that are mainly produced by tape casting. In achieving uniform, dense, dimensionally accurate ceramic sheets, the colloidal processing of tape casting slurry is the critical step. Tape casting is generally based on the non-aqueous based system, but recently the aqueous based system has gained some importance. But, each system has some advantages and disadvantages [8–12]. Non-aqueous tape casting is fast and easy to fabricate tape, but organic solvents are costly. Aqueous tape casting is low cost but it has major difficulties in drying. Success in tape casting with the required reproducibility and consistency depends on the degree of dispersion [10–12]. There are very few dispersants available for nonaqueous based tape casting such as menhaden fish oil, triton-X 100 and phosphate ester. Many reports are available on tape casting and rheological study of PZT, PLZT, and $BaTiO_3$ [13–18]. However, there are only a few studies on their electrical properties and, in particular, their piezoelectric properties

* Corresponding author.
E-mail addresses: rana_brata@hotmail.com, ranabrata_mazumder@yahoomail.com (R. Mazumder).

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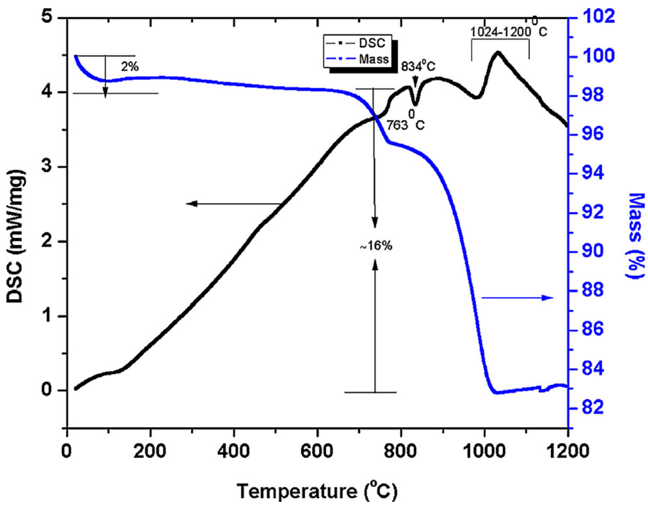

Fig. 1. Thermal analysis of mixed precursor oxides for synthesis of BZT-0.5BCT.

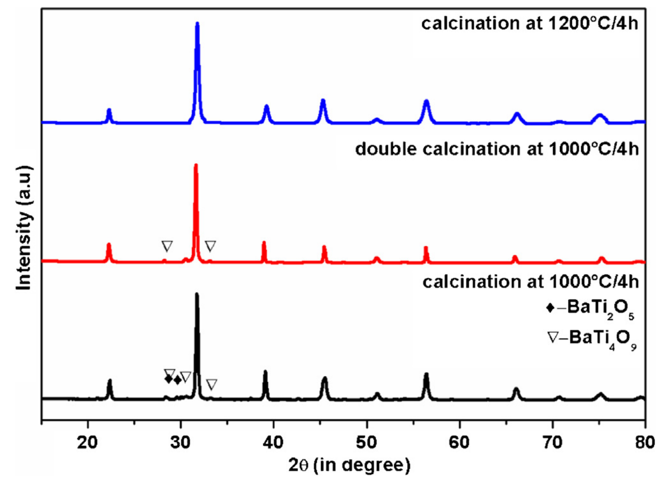

Fig. 2. X-ray diffraction patterns of BZT-0.5BCT powder calcined at different temperatures.

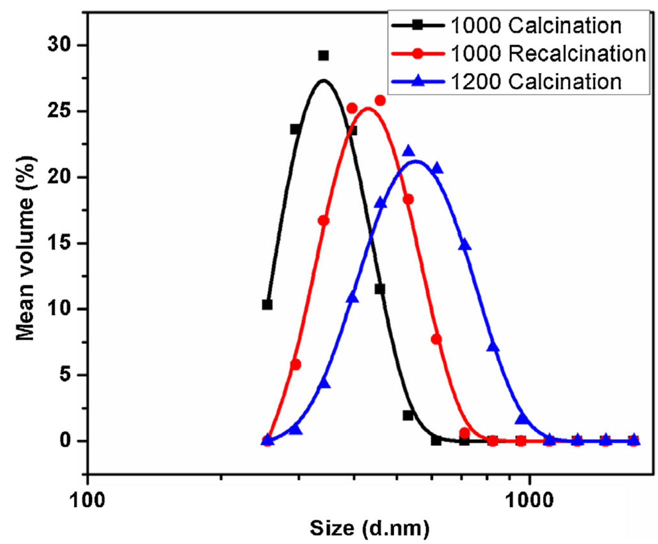

Fig. 3. Particle size distribution of calcined BZT-0.5BCT powder.

[18]. There is no report available on the fabrication of BZT-0.5BCT wafers/substrate by non-aqueous tape casting, their densification, and electrical characterization. There are many reports available on the improvement of the dielectric and piezoelectric properties of BZT-0.5BCT by varying the synthesis and the sintering conditions [19–23]. Recently, Kaushal et al. [24], studied the stability of aqueous suspensions of BZT-0.5BCT in the presence of different dispersants (dolapix, dispex A40, duramax D-3005) and they observed the Ba^{2+} and Ca^{2+} ions, leaching in aqueous media. It is obvious that aqueous tape casting may degrade the final property of piezoelectric ceramics. It has also been discussed in a few report that the dispersion of $BaTiO_3$ becomes effective if a combination of MEK-EtOH and phosphate ester is used as solvent and dispersant, respectively [13,14]. To the best of our knowledge, there is no report available on the fabrication of BZT-0.5BCT wafers/substrate by tape casting and their electrical characterization.

The present paper describes about the non aqueous tape casting of BZT-0.5BCT by studying the effect of dispersant [phosphate ester (PE)] and binder [polyvinyl butyral (PVB)] content on the slurry rheology, the properties of tape cast BZT-0.5BCT layers, its densification, dielectric and piezoelectric properties.

2. Experimental

The BZT-0.5BCT powder was prepared by solid state route. Reagent grade of barium carbonate [$BaCO_3$ (Sigma-Aldrich, 99%)], calcium carbonate [$CaCO_3$ (Sigma-Aldrich, 99%)], zirconium dioxide [ZrO_2 (Sigma-Aldrich, 99.9%)] and TiO_2 (Sigma-Aldrich, 99.0%) were used as the precursor materials. A stoichiometric amount of powders was mixed by a ball mill for 12 h with zirconia ball using isopropyl alcohol as the media. The mixture was dried at 100 °C overnight in an oven. The mixture was placed in an alumina crucible, which was subsequently inserted into the furnace and heated in the temperature range of 1000 °C–1200 °C in air.

Suspensions of BZT-0.5BCT were prepared using reagent grade solvent consisting of an azeotropic mixture of methyl ethyl ketone (MEK) and ethanol (66:34 by volume). BZT-0.5BCT and the solvent in the weight ratio of 64.9:35.1 along with 0.5–2.5 wt.% phosphate ester (Surfonic PE-1168, Richard E. Mistler Inc., USA) were milled for 4 h using zirconia balls to obtain the desired suspension. The viscosity of the slurry was measured by using a concentric cylinder rotational viscometer (VT500 Haake, Germany, equipped with the sensor system SV1) at a shear rate of 40.34 s^{-1}. Sedimentation

Table 1
Composition of BZT-0.5BCT tape casting slurry.

Ingredient	Function	Weight (%)
BZT-0.5BCT	Ceramic	65–48
Phosphate Ester (Emphos PS21-A)	Dispersant	1.0
Methyl Ethyl Ketone (E. Merck India Ltd.) + Ethanol (Bengal Chemicals & Pharmaceuticals Ltd. India)	Solvent	25–36
Polyvinyl Butyral (Hipol B-30, Hindustan Inks and Resins Ltd. Gujrat, India)	Binder	2.75–7
Polyethylene Glycol (S. D. Fine-Chem Pvt. Ltd.)	Plasticizer	3.74–5.1
Butyl Benzyl Phthalate (Merck–Schuchardt)	Plasticizer	1.5
Cyclohexanone(S. D. Fine-Chem Pvt. Ltd.)	Homogenizer	1.25