G Model
JECS-10262; No. of Pages 6
ARTICLE IN PRESS
Journal of the European Ceramic Society xxx (2015) xxx–xxx
Contents lists available at www.sciencedirect.com
Journal of the European Ceramic Society
journal homepage: www.elsevier.com/locate/jeurceramsoc

Characterization of thick bismuth ferrite–lead titanate films processed by tape casting and templated grain growth

Meghdad Palizdar[a,f,*], Chris M. Fancher[b], Tim P. Comyn[a], Tim J. Stevenson[a], Stephen F. Poterala[c], Gary L. Messing[c], Ender Suvaci[d], Annette P. Kleppe[e], Andrew J. Jephcoat[e], Andrew J. Bell[a]

[a] Institute for Materials Research, University of Leeds, LS2 9JT Leeds, UK
[b] Department of Materials Science and Engineering, North Carolina State University Raleigh, NC 27695, USA
[c] Department of Materials Science and Engineering, Pennsylvania State University, University Park, 16802, USA
[d] Department of Materials Science and Engineering, Anadolu University, Eskisehir, Turkey
[e] Diamond Light Source Ltd, Diamond House, Harwell Science and Innovation Campus, Didcot, Oxfordshire, UK
[f] R&D and Training Department, CPG Industrial, Mining and Technical Services GmbH, Dusseldorf 40237 Germany

ARTICLE INFO

Article history:
Received 12 June 2015
Received in revised form 25 August 2015
Accepted 31 August 2015
Available online xxx

Keywords:
Ferroelectric materials
Tape casting
Texture
Synchrotron

ABSTRACT

The templated grain growth technique was used to synthesise textured $60BiFeO_3$–$40PbTiO_3$ (60:40BFPT). Both Aurivillius ($Bi_4Ti_3O_{12}$, $PbBi_4Ti_4O_{15}$) and perovskite templates ($BaTiO_3$, $SrTiO_3$) were used to prepare 60:40BFPT. Only $BaTiO_3$ templates were found to successful impart a texture to the ceramic matrix. In the case of perovskite templates, ferroelectricity was evident from saturated polarisation hysteresis loops. Saturated polarisation loops were achieved due to the substitution of Ba^{2+} or Sr^{2+}, which reduces the coercive field. $SrTiO_3$ and $BaTiO_3$ templated ceramics showed remanent polarisation of 30 and 36 $\mu C/cm^2$, respectively. Aurivillius templates did not generate ferroelectric materials. Because of their high chemical stability in this system, $BaTiO_3$ templates appear to be the best candidate for fabricating textured BFPT by the reactive templated grain growth method.

1. Introduction

Materials with morphotropic phase boundaries offer the possibility of an electric field induced phase transition, for example as reported by Park and Shrout [1]. Single crystals such as $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PMN–PT) and $Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ (PZN–PT) exhibit considerable electric field induced strain [2–4]. Furthermore, Park and Shrout showed that this phase transition occurred via polarisation rotation. Complementary neutron and synchrotron X-ray diffraction on PMN–30PT and PZN–8PT revealed that this phase transition occurs through an intermediately monoclinic phase [5]. The orientation of the applied field with respect to the crystal determines the mechanism of rotation of the ferroelectric dipole [6,7].

The solid solution between bismuth ferrite and lead titanate $xBiFeO_3$-$(1-x)PbTiO_3$ or (BFPT) possesses a morphotropic phase boundary (MPB) between the rhombohedral and tetragonal forms at $x = 0.7$, with a spontaneous strain of 18% on the tetragonal side of this boundary [8–12]. The antiferromagnetic Néel temperature drops by approximately 300 K on crossing the MPB from the rhombohedral to the tetragonal side [13,14]. It is of interest to investigate the influence of field-driven rhombohedral–tetragonal phase transitions across the MPB in this system, to determine whether correctly oriented BFPT can provide both giant electric field induced strains and significant coupling with the magnetic sub-structure. A field induced transition from rhombohedral to tetragonal phases in textured BFPT could unleash strains as high as the tetragonal spontaneous strain.

Here, we used the template grain growth method (TGG) to synthesis textured BFPT. Both Aurivillius and perovskite plate-like particles were used templates. The texture of templated ceramics was determined using high-energy synchrotron.

2. Experimental procedure

Aurivillius ($Bi_4Ti_3O_{12}$, $PbBi_4Ti_4O_{15}$) and perovskite ($SrTiO_3$ and $BaTiO_3$) plate-like templates were synthesised using the molten salt method. Details of the molten salt synthesis procedure can be found in Refs. [15–19]. Templated $0.6BiFeO_3$-$0.4PbTiO_3$ (60:40 BFPT) ceramics were prepared using the reactive template grain

* Corresponding author.
E-mail address: Palizdar@cpg-gmbh.de (M. Palizdar).

http://dx.doi.org/10.1016/j.jeurceramsoc.2015.08.037

Please cite this article in press as: M. Palizdar, et al., Characterization of thick bismuth ferrite–lead titanate films processed by tape casting and templated grain growth, *J Eur Ceram Soc* (2015), http://dx.doi.org/10.1016/j.jeurceramsoc.2015.08.037

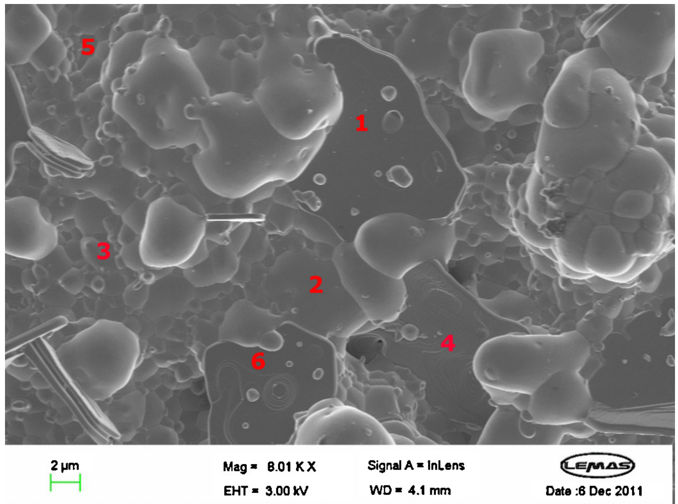

Fig. 1. SEM micrograph of the tape-cast plane for a 60$BiFeO_3$–40$PbTiO_3$ ceramic templated with 10 wt.% $BaTiO_3$.

growth method. The casting slurry was prepared using the BFPT precursor powder plus 10% by weight of template ($Bi_4Ti_3O_{12}$, $PbBi_4Ti_4O_{15}$, $SrTiO_3$ and $BaTiO_3$), an azeotropic ratio of solvents, binder, plasticizers and dispersant. The slurry was casting by using a shear rate of 512 s^{-1}, achieved by a combination of doctor blade height and speed [16,17,20]. After drying, BFPT templated with $Bi_4Ti_3O_{12}$, $PbBi_4Ti_4O_{15}$, or $SrTiO_3$ were sintered at 1050 °C for 1 h and BFPT templated with $BaTiO_3$ was sintered at 1100 °C for 1 h, respectively. The obtained layers are between 0.8 and 1.2 mm thick. To compare the influence of different types of templates on the chemical and physical properties of the material, 60:40 BFPT, using no template, was synthesised as a reference via tape casting and was sintered at 1100 °C for 1 h.

Scanning electron microscopy (SEM, Zeiss Leo FEG-SEM, Carl Zeiss SMT Ltd.), was used to characterize the grain morphology of templated BFPT. Strain and polarisation measurements where measured using a Radiant LC precision ferroelectric characterization instrument. Dielectric measurements of the sample were carried out using a HP4192A LF impedance analyzer. Synchrotron diffraction was carried out at beam line I15 at the Diamond Light Source facility (Oxfordshire, UK) using hard synchrotron radiation ($\lambda \approx 0.216$ Å). Diffraction data was measured using a 2D image plate (MAR 345). Measured Debye rings were "caked" into individual 2θ-intensity diffraction patterns, at +/− 5° between $\psi = 0°$ and 355°.

3. Results and discussion

3.1. Texture analysis

Table 1 summarises the dielectric and crystallographic characteristics of 60:40 BFPT synthesised with different templates. It reveals that among those templates, $BaTiO_3$ was able to impose the texture morphology to the matrix, partially. However, there are some drawbacks, such as reduction in Curie temperature (T_c) and *c/a* ratio.

Chemical compositions in Table 1 have been calculated from average EDX spot scan analysis on some parts of samples, ignoring the oxygen content. The SEM micrograph of 60:40BFPT templated with $BaTiO_3$ (Fig. 1) shows a dense microstructure, which consists of both spherical and plate-like regions. The plate-like regions

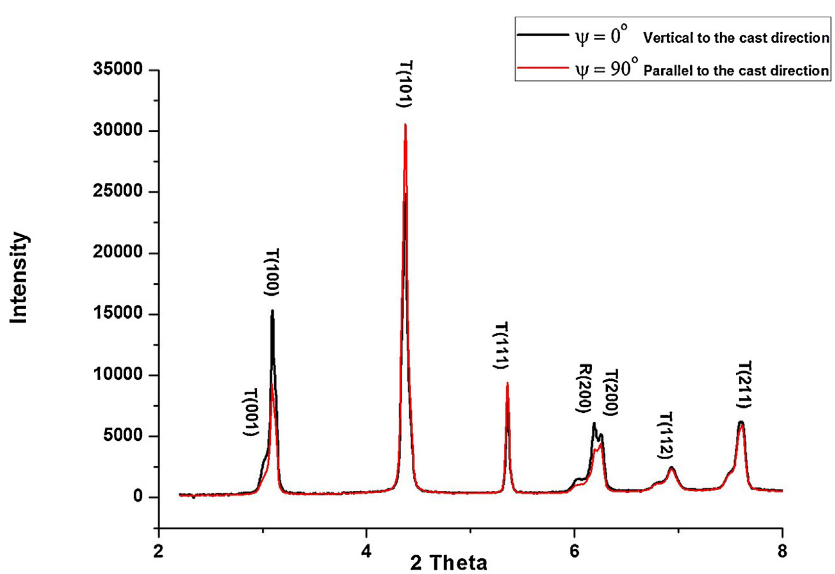

Fig. 2. Comparison of synchrotron XRD patterns for 60:40BFPT made from TGG method using 10% $BaTiO_3$ for a scattering vector parallel to the sample normal ($\psi = 0°$) and parallel to the cast direction ($\psi = 90°$).