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TEMPLATED GRAIN GROWTH - THE ART AND SCIENCE OF PATTERNING CERAMIC MICROSTRUCTURES

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The Templated Grain Growth (TGG) process is a relatively new process to texture ceramics. Three ferroelectric ceramic systems are used to illustrate the factors controlling texture development during TGG. TGG was utilized to fiber-texture both $(Sr_{0.53}Ba_{0.47})Nb_2O_6$*, and* $Pb(Mg_{1/3}Nb_{2/3})O_3$*-*$PbTiO_3$*, in the [001] and to sheet-texture* $(Sr_{0.90}La_{0.10})_2Nb_2O_7$*. A direct correlation between crystallographic texture and properties is displayed for all three systems. An enhancement of either the piezoelectric, pyroelectric, and/or dielectric response due to the crystallographic texturing in the desired direction is evident for all systems.*

INTRODUCTION

Traditional bulk ceramic processing has usually resulted in the formation of microstructures which exhibit physical properties governed by the $\infty\infty m$ symmetry. Each grain in the polycrystalline sample has a crystallographic orientation different from the neighboring grains. This random distribution results in an averaging of properties in all directions, often leading to an isotropic appearance for even an anisotropic system which usually displays anisotropic properties in single crystal form. In contrast, textured ceramics can exhibit anisotropic physical properties which are typical of single crystals. Therefore, enhanced and anisotropic electrical, electromechanical, magnetic, optical, and structural properties can be achieved in a ceramic sample with the desired texture.

TGG offers the possibility of fabricating grain-oriented polycrystalline ceramics which exhibit single crystal-like properties in the desired crystallographic direction. It has been shown that oriented template particles can be used to develop textured microstructures by TGG in alumina[1], mullite[2], and $Bi_4Ti_3O_{12}$[3]. The process involves the orientation of anisotropically-shaped template particles in a dense, fine-grained matrix by shear-forming techniques. Template particles must be large and anisometric in shape, so that they can be oriented during forming and grow preferentially during heating. The driving force for the TGG process is the difference in surface free energy between the templates and the matrix grains. The relative size of the matrix and template particles during densification plays an important role in texture development[4,5]. The final texture in the microstructure depends strongly on the initial number of template particles and their distribution throughout the body.

In this paper, we report on the texturing of three ferroelectric systems, $(Sr_{0.53}Ba_{0.47})Nb_2O_6$, $(Sr_{0.90}La_{0.10})_2Nb_2O_7$, and $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$. We were interested in texturing the first two ceramic systems in the spontaneous polarization direction for the benefit of poling efficiency and increased properties. $(Sr_xBa_{1-x})Nb_2O_6$ possesses the tetragonal tungsten bronze structure for x=0.25-0.75[6] which shows either normal or relaxor ferroelectric properties, depending on the composition[7]. For this system, we were interested in fiber-texturing (1-D texturing) the ceramic in the [001] utilizing acicular $KSr_2Nb_5O_{15}$ particles. $(Sr_{0.90}La_{0.10})_2Nb_2O_7$ is a regular ferroelectric material with a perovskite-layered structure ($Ca_2Nb_2O_7$ structure)[8]. In this system, the required texturing direction is along the c-axis or [001], which was achieved by using blade-like $Sr_2Nb_2O_7$ templates. $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) is different than the above systems. PMN-PT is a perovskite relaxor ferroelectric material which displays a rhombohedral (or pseudo-cubic) symmetry at a composition below the morphotropic phase boundary (<0.35 mol% $PbTiO_3$)[9]. Fiber-texturing in the [001] is also required for this system even though the spontaneous polarization is in the <111>. Park *et al* showed that PMN-PT compositions just below the MPB (30-34 mol% $PbTiO_3$) displayed enhanced piezoelectric response in the <001> under high field conditions due to a phase transformation from the rhombohedral to the tetragonal state. Therefore, it is desirable to fiber-texture the PMN-PT composition near the MPB in the <001>, and this was accomplished by using oriented {001}-$BaTiO_3$ tabular particles.

This study focuses on the texturing of these three ferroelectric systems by TGG, and it shows the effect of texturing on the piezoelectric, pyroelectric, and/or dielectric properties. The properties achieved were compared and contrasted to single crystal and randomly oriented ceramic properties for the three systems.

RESULTS AND DISCUSSION

Fiber-Textured $(Sr_{0.53}Ba_{0.47})Nb_2O_6$

Textured $(Sr_{0.53}Ba_{0.47})Nb_2O_6$ (SBN53) ceramics with a relative density >95% were fabricated by TGG[10]. 0.85 mol% V_2O_5 was used as a liquid former to enhance the grain growth of the SBN53 ceramic grains. $KSr_2Nb_5O_{15}$ template particles, synthesized by a molten salt process, were aligned by tape casting in a mixture of solid-state-synthesized $SrNb_2O_6$ and $BaNb_2O_6$ powders. The resulting ceramics possess strong fiber-texture along the polar axis, [001], of SBN53. Samples with 15.4 wt% templates reached a texture percentage >90% after sintering at 1200 °C for 4 h (Figures 1, 2). These samples showed a peak dielectric constant of 7550 (at 1 kHz) at the Curie temperature (145 °C), and a room temperature remanent polarization of 13.2 $\mu C/cm^2$ and saturation polarization of 21 $\mu C/cm^2$ (60-85% of single crystal)[11], piezoelectric strain coefficients of 78 pC/N (70-85% of single crystal)[11, 12], and pyroelectric coefficients of 2.9×10^{-2} $\mu C/cm^2$ °C (52% of single crystal)[13]. These results show that TGG is a viable option for accessing single crystal properties in polycrystalline ceramics.

Sheet-Textured $(Sr_{0.90}La_{0.10})_2Nb_2O_7$

$(Sr_{0.90}La_{0.10})_2Nb_2O_7$ (SLN) ceramics reached approximately 97% of theoretical density when sintered between 1450-1500 °C for 1-4 h[14]. These ceramics were sintered with a minimum amount of excess niobium (500-1000 ppm) to produce the required liquid phase, which enhanced the growth of the aligned template grains, 10 vol% $Sr_2Nb_2O_7$ (SN) blade-like templates,

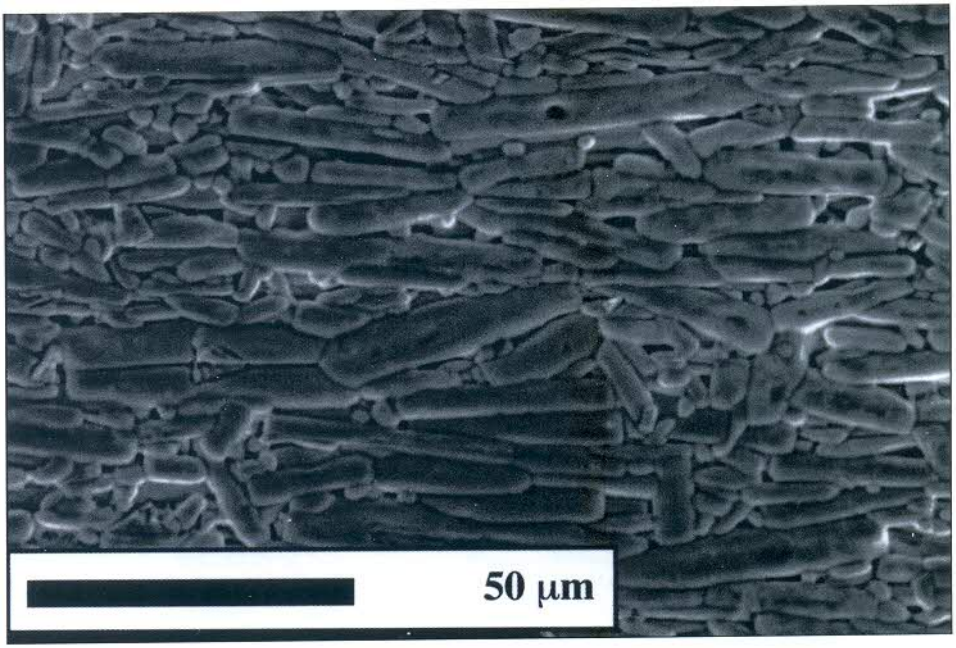

FIGURE 1 - SBN53 textured in the [001] with 15.4 wt% $KSr_2Nb_5O_{15}$ templates + 0.85 mol% V_2O_5 sintered at 1350 °C for 4 h.

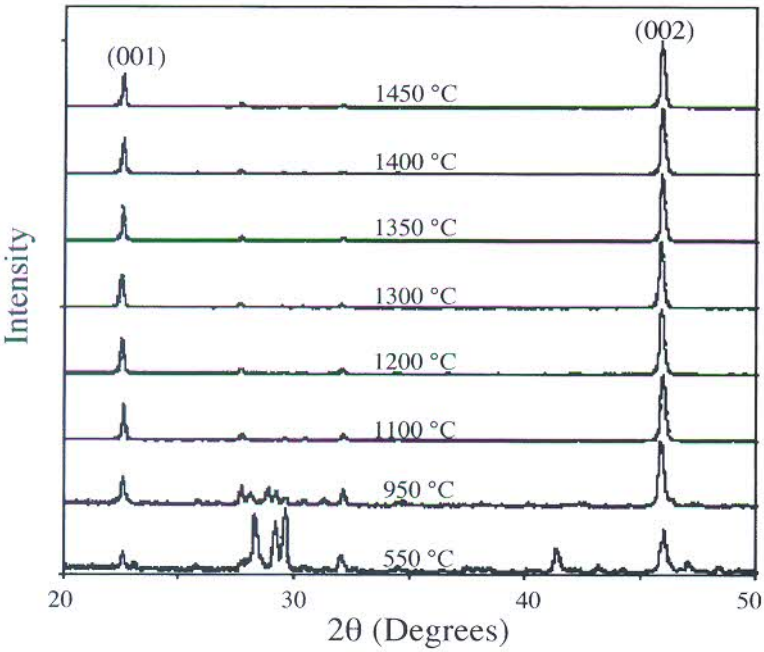

FIGURE 2 - Texture evolution of (Sr,Ba)Nb_2O_6 containing 15.4 wt% $KSr_2Nb_5O_{15}$ templates + 0.85 mol% V_2O_5 sintered at 1400 °C for 4 h.

formed by molten salt synthesis in KCl[4], were initially aligned in the SLN matrix powder by tape casting through 1.0 millimeter-spaced gates located behind the doctor blade. The gates provided the required shear field to orient the length of the templates along the casting direction. This tape casting process made the b-planes of all the templates parallel to each other and the tapecast surface, and made a particular direction (a-direction) in the oriented b-plane of all templates parallel to a common direction (casting direction). By achieving this alignment of the blade-like templates, the proper initial green processing conditions for sheet-texture were satisfied, and thus, the texture was developed with further heat treatment during sintering.

TGG samples of SLN containing 10 vol% blade-like SN templates sintered at 1450 °C for 4 h displayed distinct sheet-texture (3-D texturing) when characterized by XRD (Figure 3, 4). Along the tape casting direction (a-direction), the (200) was the most prevalent plane, while in the polarization direction (c-direction) and the tape face direction (b-direction) displayed the (002) and (080), respectively. The dielectric behavior of the sheet-textured TGG samples was measured in the temperature range of -150 °C to 450 °C at 1 MHz for all orthogonal directions. The room temperature dielectric behavior for the a-,

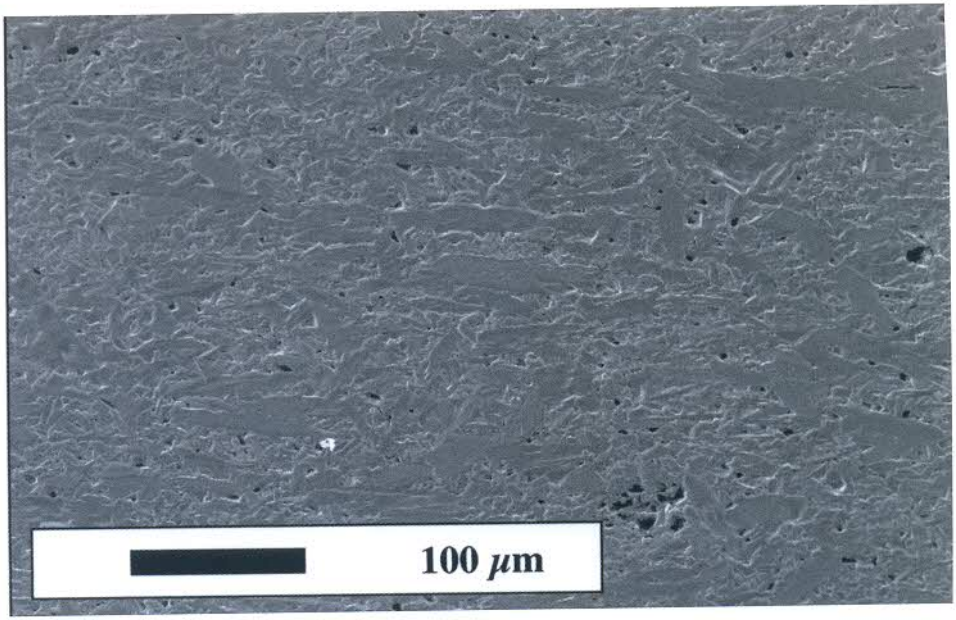

FIGURE 3 - (Sr,La)$_2$Nb$_2$O$_7$ textured in the [001] with 10 vol% Sr$_2$Nb$_2$O$_7$ templates sintered at 1450 °C for 1 h.

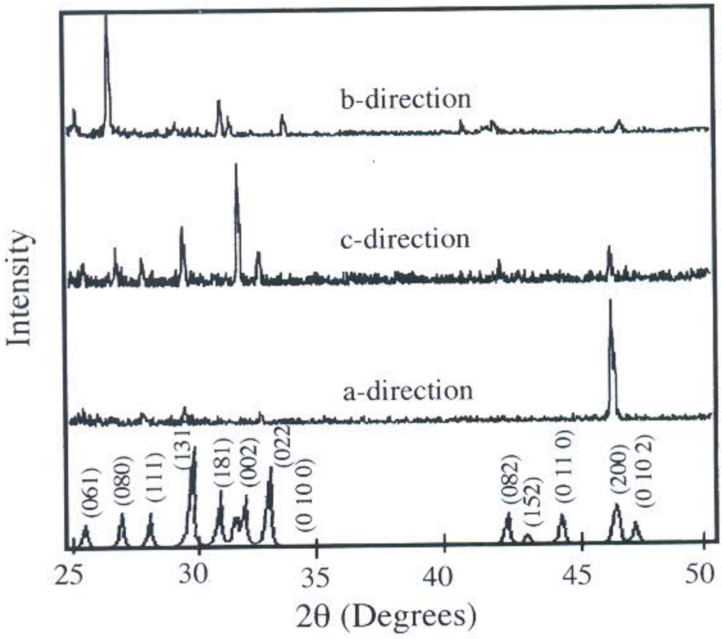

FIGURE 4 - Directional dependence of (Sr,La)$_2$Nb$_2$O$_7$ textured with 10 vol% Sr$_2$Nb$_2$O$_7$ templates sintered at 1450 °C for 1 h.