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X ray investigation of high oriented $(1-x)PbMg_{1/3}Nb_{2/3}O_3$–$(x)PbTiO_3$ ceramics

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Abstract

Textured Tape Cast Ceramics were prepared by homo epitaxial template grain growth from $(1-x)PbMg_{1/3}Nb_{2/3}O_3$–$(x)PbTiO_3$ (PMN–PT) particles and cubic templates. The texture fraction of (0 0 1) oriented ceramics was estimated by the Lotgering method at RT, in the ferroelectric phase, and above T_c, in the cubic phase. Textured Tape Cast Ceramics display quasi complete (0 0 1) texture ($f = 0.99$). The (0 1 1) pole figure reveals (0 0 1) fibre texture and small orientation distribution.

Keyword: Relaxor materials

1. Introduction

Orientation dependent of single crystals properties lead to the prediction that ceramics exhibiting high degrees of appropriate crystallographic texture should have an electromechanical properties approaching those of [0 0 1] oriented single crystals. $(1-x)PbMg_{1/3}Nb_{2/3}O_3$–$(x)PbTiO_3$ (PMN–PT) ceramics, with the high texture fraction were obtained by reactive template grain growth using Sr/$BaTiO_3$ templates.[1,2] Although very high texture was archived, piezoelectric properties of textured ceramics are far from those obtained for single crystal. Porosity, micro-cracks and [0 0 1] large orientation distribution are the factors that could affect and restrict the piezoelectric response of the textured samples.

This paper reports the estimation of texture fraction in the cubic phase using Lotgering method of oriented PMN–PT ceramics obtained by homo epitaxial template grain growth from PMN–PT particles and cubic templates. Pole figure and rocking curve of PMN–PT textured ceramics were recorded.

2. Experimental methods

PMN–PT textured ceramics were synthesized by homo-epitaxial template grain growth from PMN–35%PT nano powders and large PMN–25%PT cubic template via tape casting.[3]

An advanced study of the texture is obtained using a high accuracy two-axes diffractometer using Cu Kα monochromatic radiation issued from a Rigaku rotating anode (RU 300, 18 kW)[4]: ceramics were fixed on a sample, mounted upon a HUBER goniometric head. A N_2 flow cryostat was used to record spectra at 473 K in the cubic phase.

The texture fraction of the {0 0 1} planes was estimated from the XRD patterns using the Lotgering method. The Lotgering factor f is defined as the fraction of area textured with the crystallographic plane of interest[5] using the formula

$$f_{00l} = \frac{P_{00l} - P_0}{1 - P_0}$$

where

$$P_{00l} = \frac{\sum I_{00l}}{\sum I_{hkl}}$$

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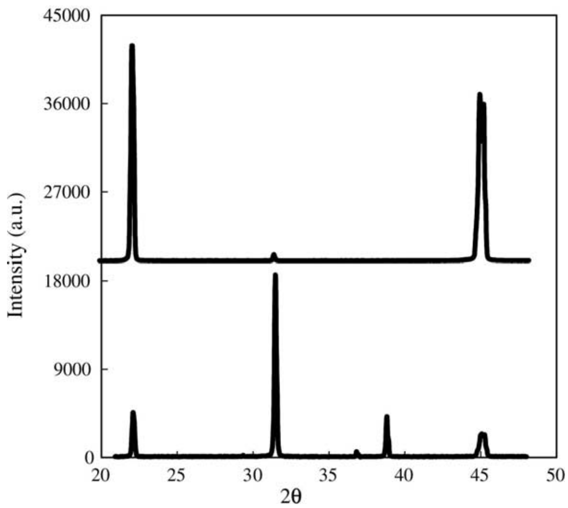


Fig. 1. Diffraction X-ray spectra of PMN–PT random oriented ceramic (bottom) and Textured Tape Cast Ceramics (top).

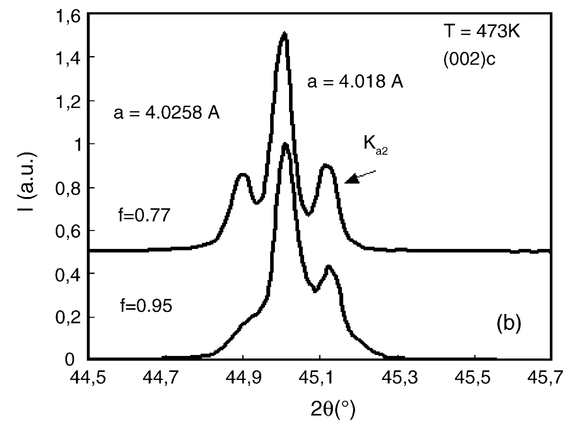


Fig. 2. Diffraction X-ray spectra of {0 0 2} at 473 K of samples displaying the Lotgering factors of 0.77 and 0.95.

and

$$P_0 = \frac{\sum I^0_{00l}}{\sum I^0_{hkl}}$$

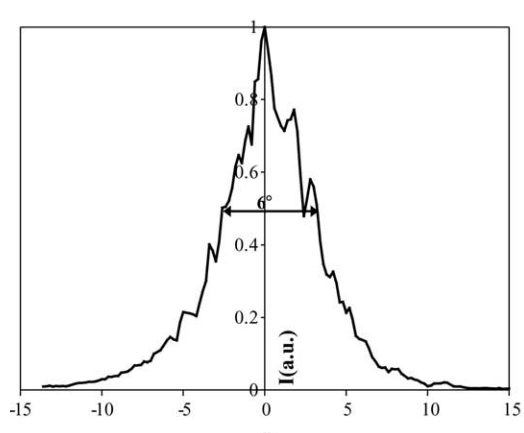
with I_{hkl} and I^0_{hkl} the intensities of $(h\,k\,l)$ peaks for the textured and for the randomly oriented sample, respectively. It is important to note that the analysed surface is about $\sim$1 cm^2 and the penetration deep of X-ray at 20–40 keV is few microns. Furthermore, textured ceramics exhibit grains size of about 50–200 μm.

3. Results and discussion

XRD spectra of random orientation ceramic show the highest intensity for the (1 1 0) peak. For Textured Tape Cast Ceramics (TTCC) samples, the intensity of the (1 1 0) peak decreases and a strong increase of the $(0\,0\,l)$ diffraction peak intensities was observed (Fig. 1). This intensity variation indicates an increase in the crystallographic $(0\,0\,l)$ orientation.

The texture fraction or Lotgering factor of textured samples was estimated from XRD spectrum recorded at RT using all peaks up to (2 2 2), i.e. 2θ up to 85°. For ceramics sintered at 1150 °C and at 1200 °C for 10 h, the TTCC disks ($\Phi \sim 14$ mm) exhibit texture fraction of ∼0.77 (sample A) and ∼0.95 (sample B), respectively. Above Curie temperature, at 473 K, the (0 0 2) reflection peaks become narrow and shift towards low value. A doublet was observed near 44.9° and 45° instead of only one in the cubic symmetry. The two peaks were thus related to the two different compositions. The corresponding a parameters are equal to 4.026 and 4.018 Å, respectively. The most important peak (4.018 Å) corresponds obviously to the PMN–35%PT since it accounts for approximately ∼70% volume of the matrix for sample A. The second one is related to PMN–25%PT cubic templates. On the other hand, the peak corresponding to cubic templates which is well observed for sample A, is very weak and observed as a shoulder for sample B displaying a texture fraction higher than 0.9 (Fig. 2).

These results show that for TTCC disks, the two compounds PMN–25%PT and PMN–35%PT can be distinguished in the cubic phases. For better estimation of the



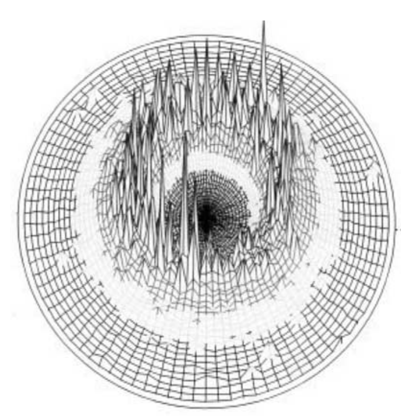


Fig. 3. ω-scan of the {0 0 2} peak, recorded at RT on the as-sintered surface of sample displaying the Lotgering factor of 0.95 (left) and the {0 1 1} pole figure of a textured ceramic.

texture fraction, the Lotgering factor *f* was then calculated from XRD spectra recorded at 473 K in the cubic phase. The factor *f*, determined in the cubic phase, is higher than that one calculated from XRD at room temperature. The calculated Lotgering factors are 0.9 in the cubic (or paraelectric phase) instead of 0.6 in the RT (or ferroelectric phase) for sample A. For quasi-complete tape cast textured ceramic (sample B), the factors *f* is 0.99 in the cubic phase, very close to that calculated at RT (0.95).

The two textures observed on the XRD spectra of textured samples in the cubic phase reveals that the composition of the grown oriented grains is determined by the matrix composition. The presence of two textured compositions in PMN–PT Tape Cast Textured Ceramics is consistent with ceramic texturing through the liquid phase at the crystal–matrix interface.

The orientation relationship between grains was investigated by a ω-scan of the {0 0 2} reflection and (0 1 1) pole figure. Fig. 3 shows the ω-scan of the {0 0 2} peak, recorded at RT, on as sintered surface. The ω-scan of the {0 0 1} peak recorded on polished surface present similar curves. The full width at half maximum (FWHM) of the peak is equal to 9° and 6° for samples displaying texture of 0.7 and 0.95, respectively. For the last sample, the Lotgering factors, calculated from XRD spectra recorded in the Bragg–Brentano geometry in symmetric configuration and along texture axis configuration, are 0.95 and 0.96, respectively. Therefore, we can conclude that orientation distribution of {0 0 *l*} direction in textured ceramics is very small and out of plan alignment is good. The ring pattern (Fig. 3) of the highest contour is characteristic of the fibber texture, i.e. with no preferential orientation in the plane.

4. Conclusion

This work shows that the quasi complete ⟨0 0 1⟩ texture of PMN–35%PT ceramic was obtained ($f > 0.95$). Even for this high textured ceramic, only fibre texture showing small orientation distribution was obtained. The two textures observed by X-ray diffraction indicate that the grown textured ceramic is determined by the matrix composition. Therefore, it is not necessary that the composition of the pristine seed dispersed in the green matrix have the target composition.

Acknowledgments

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