Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Enhanced piezoelectric properties and temperature-insensitive strain behavior of < 001 > -textured KNN-based ceramics

Baihui Liu, Peng Li, Bo Shen, Jiwei Zhai*, Yang Zhang, Feng Li, Xing Liu

Key Laboratory of Advanced Civil Engineering Materials of Ministry of Education, School of Materials Science & Engineering, Tongji University, Shanghai 201804, China

ARTICLE INFO

Keywords:
KNN-based textured ceramics
Microstructure
Piezoelectric properties
Temperature stability

ABSTRACT

In this study, < 001 > -textured $0.99(K_{0.5}Na_{0.5})_{0.95}Li_{0.05}Nb_{0.93}Sb_{0.07}O_3$–$0.01CaZrO_3$ [abbreviated as 0.99KNLNS-0.01CZ] lead-free ceramics were prepared by templated grain growth (TGG) using plate-like $NaNbO_3$ templates and sintered by a two-step sintering process with different soaking time. All textured samples with high Lotgering factor (f > 85%) presented orthorhombic and tetragonal coexisting phase, and the proportion of orthorhombic phase was varied with prolonged soaking time. A large piezoelectric constant d_{33} (~ 310 pC/N) was obtained in the textured samples with a 12 h soaking time, which was almost twice larger compared to the randomly oriented one. Furthermore, the field-induced piezoelectric strain coefficient d_{33}^*(~ 440 pm/V) of the textured ceramics with 6 h soaking time was larger than the value of randomly oriented one (~ 298 pm/V) at room-temperature. Enhanced piezoelectric response and good temperature stability prove that < 001 > -textured 0.99KNLNS-0.01CZ ceramics are promising candidates in the field of lead-free piezoelectric materials.

1. Introduction

It's well known that the piezoresponses of single crystals with perovskite structure usually present an anisotropy characteristic [1]. It has been reported in Spark's work that the longitudinal piezoresponse d_{33} along the non-polar $< 001 >_{pc}$ direction of PMN-PT single crystal was 2500 pC/N, thirteen times higher than the value (190 pC/N) of < 111 > poled one [2]. However, the preparation technique of single crystals is complex and directional cutting is extremely difficult [3]. Therefore, since Satio and his co-workers achieved excellent piezoelectric properties (d_{33} =416 pC/N, k_p =0.61) in < 001 > -oriented KNN-based piezoelectric ceramics using TGG method in 2004 [4], the lead-free textured ceramics have attracted great attention both in preparation process and performance.

Utilizing TGG method, plate-like templates are aligned towards a preferred direction and the epitaxial growth of matrix driven by the difference of surface free energy between the matrix and template particles is on the surface of templates [5]. After the sintering and annealing process, textured ceramics can be successfully fabricated and the piezoresponse of textured ceramics usually show an anisotropy nature, which is similar to the single crystals. Over the past decades, enhanced piezoelectric properties in KNN-based ceramics have been achieved by applying TGG technique. Chang *et al.* systematically studied < 001 > textured $(K_{0.5}Na_{0.5})_{0.98}Li_{0.02}NbO_3$, $(K_{0.5}Na_{0.5})(Nb_{0.85}Ta_{0.15})O_3$ [6], CuO-doped $(K_{0.476}Na_{0.524})NbO_3$ [5] and $(K_{0.5}Na_{0.5})Nb_{0.97}Sb_{0.03}O_3$ [7] lead-free ceramics and obtained improved piezoelectric properties than their randomly oriented equivalents. Satio's group concentrated on using different kinds of templates to prepare textured pure KNN ceramics [8,9], however, the improvement of piezoelectric properties were limited due to the restriction of compositions. The improvement of the longitudinal piezoresponse for KNN-based ceramics by texture development is about 20–70% according to previous study, while the improvement of lead-based ceramics is 100% or higher [10]. According to Yan's research, d_{33} of < 001 > -textured $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ ceramics could reach up to 1000 pC/N, which was about two times higher than the value of its randomly oriented ceramics [11]. Therefore vast effects still need to be concentrated on textured KNN-based ceramics.

In this study, the phase structure and microstructure as well as piezoelectric, ferroelectric and dielectric properties of < 001 > -textured $0.99(K_{0.5}Na_{0.5})_{0.95}Li_{0.05}Nb_{0.93}Sb_{0.07}O_3$–$0.01CaZrO_3$ ceramics are systematic studied and compared with its randomly oriented specimens. It can be found that textured 0.99KNLNS-0.01CZ ceramics display better performance and temperature stability than the randomly oriented one. What' more, soaking time have a great effect on the properties of textured ceramics.

* Corresponding author.
E-mail address: apzhai@tongji.edu.cn (J. Zhai).

http://dx.doi.org/10.1016/j.ceramint.2017.03.090
Received 13 February 2017; Received in revised form 15 March 2017; Accepted 15 March 2017

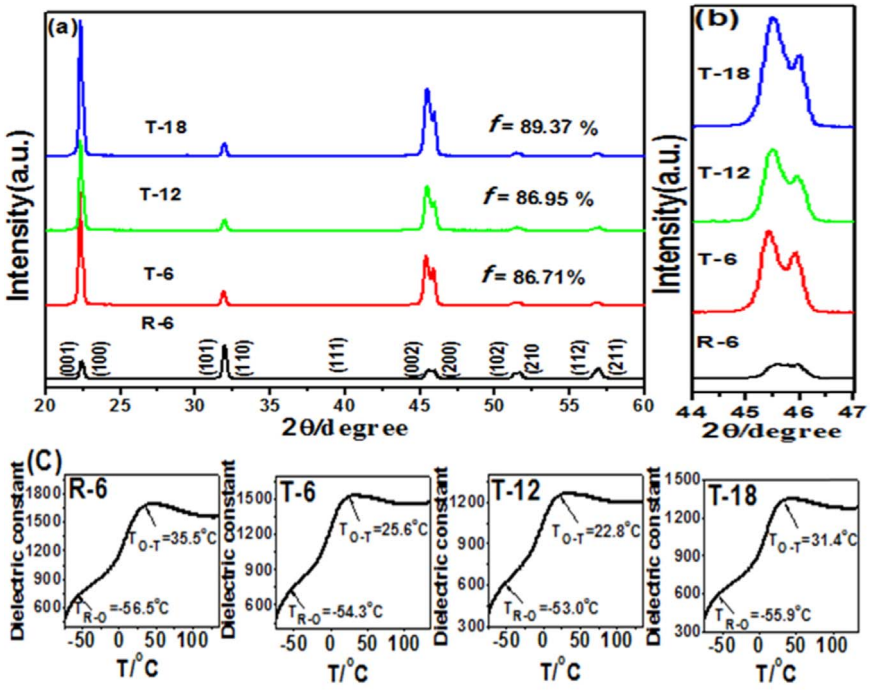

Fig. 1. (a) XRD patterns of the 0.99KNLNS-0.01CZ ceramics; (b) enlarged XRD patterns in the range of 2θ from 44° to 47°; (c) cryogenic ε_r-T curves of ceramics measured from −75 to 130 °C at 10 kHz.

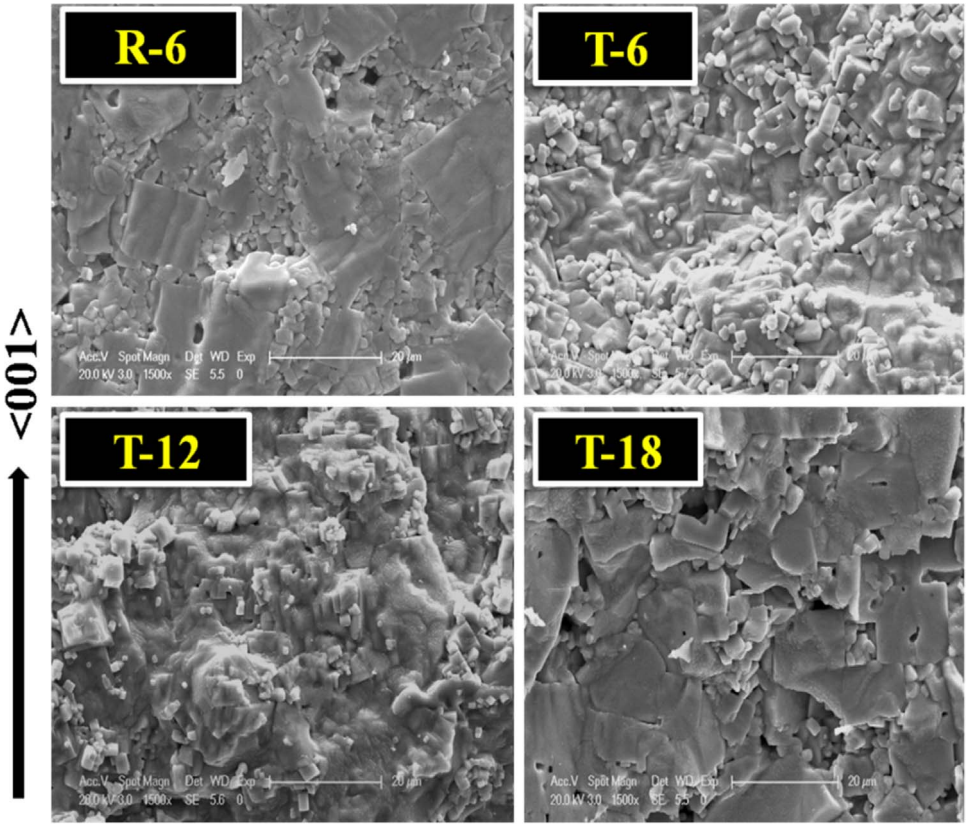

Fig. 2. Thermally etched cross-section SEM images of 0.99KNLNS-0.01CZ ceramics.

2. Experimental procedure

Textured lead-free 0.99KNLNS-0.01CZ ceramics were prepared by TGG method using 3 mol% $NaNbO_3$ (NN) templates. Plate-like NN templates with a high aspect ratio (average 15 μm in length and 5 μm in width) were synthesized by two-step molten salt method. 0.99KNLNS-0.01CZ matrix powders were synthesized by conventional solid state reaction process utilizing Na_2CO_3 (99.95–100.05%), K_2CO_3 (99%), Li_2CO_3 (99%), Nb_2O_5 (99%), Sb_2O_5 (99.998%), ZrO_2 (99%), $CaCO_3$ (99.95–100.05%) as the raw materials. The matrix and templates were mixed and roller milled to obtain homogeneous slurry. Then tape casting was performed to achieve thin tapes. Dried tapes