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Dielectric and piezoelectric properties of sodium potassium niobate-based ceramics sintered in microwave furnace

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HIGHLIGHTS

- In this work KNN-based ceramics modified with $SrTiO_3$ were fabricated.
- Sintering was carried out in microwave furnace and conventional furnace.
- Microwave sintering resulted in ceramics with higher density and more uniform microstructure.
- The d_{33} of microwave sintered samples were measured more than 50% higher than those sintered via other method.
- By addition $SrTiO_3$ higher than 3 mol%, ceramics showed relaxor behavior.

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ABSTRACT

In this work $K_{0.48}Na_{0.48}Li_{0.04}Nb_{0.96}Ta_{0.04}O_3$ ceramics modified with different amounts of $SrTiO_3$ (from 0 to 10 mol %) were fabricated via conventional sintering as well as microwave sintering. Microstructure, dielectric and piezoelectric properties of sintered ceramics via both methods, were investigated and compared. Microstructure of samples revealed that microwave sintering results in denser, more fine and uniform microstructure compared to conventional sintering. Ceramics containing more than 3% $SrTiO_3$ showed relaxor behavior. Moreover microwave sintering enhanced relaxor behavior. Since ceramics containing 1% $SrTiO_3$ had orthorhombic and tetragonal structures simultaneously, these ceramics showed maximum piezoelectric constant. Moreover microwave sintered ceramics due to denser as well as closer composition to stoichiometry, reveled higher piezoelectric constant compared to conventional sintered ones.

1. Introduction

Piezoelectric ceramics because of their electromechanical behavior are used in many applications such as sensor, actuator, transducer and generator [1]. An important group of lead-free piezoceramics is based on sodium potassium niobate (KNN) with relative good piezoelectric properties and high Curie temperature. One of difficulties through fabrication KNN ceramics which has been postponed their replacement instead of lead-based piezoceramics, is their low sinterability and possibility of alkali elements loss during sintering at high temperatures. Obtaining not enough density at low sintering temperatures and the risk of volatilization of sodium and potassium during high temperature sintering, reduce reproducibility and lower properties of fabricated ceramics [2–4].

Microwave sintering of some ceramic materials such as Al_2O_3 [5], ZnO [6,7] and ZrO_2 [8] accompanied with reduction in sintering temperature as well as sintering time compared to conventional sintering. Literature survey revealed that there is not so many published work about using microwave for sintering KNN-based compounds. However it can be found many papers which report microwave sintering of lead-based [9–12] and even lead-free based such as $BaTiO_3$ [13,14] piezoelectric compounds. In these papers improvement of density as well as electric properties of microwave sintered ceramics compared to conventional sintered ones have been reported.

In this work, $K_{0.48}Na_{0.48}Li_{0.04}Nb_{0.96}Ta_{0.04}O_3$ (abbreviated as KNNLT) was selected as based composition and the effect of

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addition different amounts of $SrTiO_3$ (abbreviated as ST) from 0 to 10 mol% on phase structure and properties were investigated. Fabricated powder compounds were sintered via two different methods: conventional furnace sintering (CFS) and microwave sintering (MWS). Microstructure, dielectric properties and piezoelectric properties of CFSed and MWSed samples were studied and compared.

2. Experimental procedure

$(1-x)$KNNLT $-$ xST $(0.0 \leq x \leq 0.10)$ powders were synthesized by a conventional solid state reaction method using Na_2CO_3, K_2CO_3, Li_2CO_3, Nb_2O_5, Ta_2O_5, SrO and TiO_2 with more than 99% purity as raw materials. At first, these powders were dried in oven at 100 °C for 24 h and then were weighed for each composition according to its stoichiometry formula. Each powder mixture was ball-milled for 24 h with zirconia balls using ethanol media. The dried slurries were calcined at 880 °C for 6 h. In order to increase composition homogeneity, the calcination was carried out twice separating by 24 h ball milling. Calcined powders were mixed with poly vinyl alcohol as a binder and then pressed into green disks with a diameter of 12 mm at 50 MPa. Green disks were sintered via conventional furnace sintering (CFS) as well as microwave sintering (MWS). A multimode 2.45 GHz microwave furnace with a power output of 3 kW was used in the MWS process. The Archimedes method was used to measure the density of sintered ceramics and the temperature that yielded the maximum relative density for each composition, was selected as the optimum sintering temperature. The microstructure of ceramics was observed using scanning electron microscopy (FE-SEM, JEOL, JSM-65OFF, Japan). The crystal structure of samples was determined using an X-ray diffractometer (XRD, X'pert PRO MRD, Philips). For electrical measurements, silver paste was applied on the lapped surfaces of the disks to serve as the electrodes. The temperature dependence of dielectric properties was measured using an impedance analyzer (HP 4192A) in a temperature range of 25–500 °C. Some samples were poled at 100 °C in silicon oil bath by applying an electric field of 3–4 kV/mm for 30 min. The piezoelectric constant (d_{33}) was measured using a piezo-d_{33} meter (ZJ-6B, China).

3. Results and discussion

3.1. Structure and phase determination

X-ray diffraction (XRD) patterns of $(1-x)$ KNNLT $-$ xST ceramics sintered at optimum sintering temperature via CFS are shown in Fig. 1. By comparing XRD patterns with JCPDS pattern 01-071-2171 (corresponding to orthorhombic $KNbO_3$) and 01-071-0945 (corresponding to tetragonal $KNbO_3$) it can be recognized that all samples show pure perovskite structure without any secondary phase, indicating formation of solid solution between KNNLT and ST within the studied range $0.0 \leq x \leq 0.10$. Crystalline type of perovskite structure is usually determined based on the relative intensities of split peaks around $2\theta = 46°$. Therefore the magnified XRD patterns of ceramics in the range $44° < 2\theta < 47°$ are shown in Fig. 2. It can be seen that XRD patterns of ceramic with $x = 0.0$ shows (202)/(020) peak splitting around $2\theta = 46°$, confirming the orthorhombic symmetry. This observation is in good agreement with reported results by Guo et al. [15]. XRD patterns of ceramic with $x = 0.0$ shows (202)/(020) peak splitting around $2\theta = 46°$, confirming the orthorhombic symmetry. For ceramics with $0.03 \leq x \leq 0.075$, XRD patterns show (002)/(200) peak splitting, indicating the tetragonal symmetry. The split peaks around $2\theta = 46°$ in XRD pattern of ceramic with $x = 0.01$ have equal intensities which is due to the coexistence of orthorhombic and tetragonal structures. The merging the split peaks together for ceramic with $x = 0.10$ indicates formation of pseudocubic structure for this composition. By comparing the peak angles of different compositions, it can be found that with increasing ST amount, the diffraction peaks shift toward higher angles, indicating the shrinkage of unit cell. This result seems to be due to the smaller ionic radius of Sr^{2+} ions (1.44 Å, CN = 12) compared to the average ionic radius of A-site matrix ions, i.e. Na^+ (1.39 Å, CN = 12) and K^+ (1.64 Å, CN = 12), as well as the smaller Ti^{4+} ion (0.61 Å, CN = 6) compared to Nb^{5+} (0.64 Å, CN = 6) which is the matrix B-site ion in the ABO_3 perovskite structure.

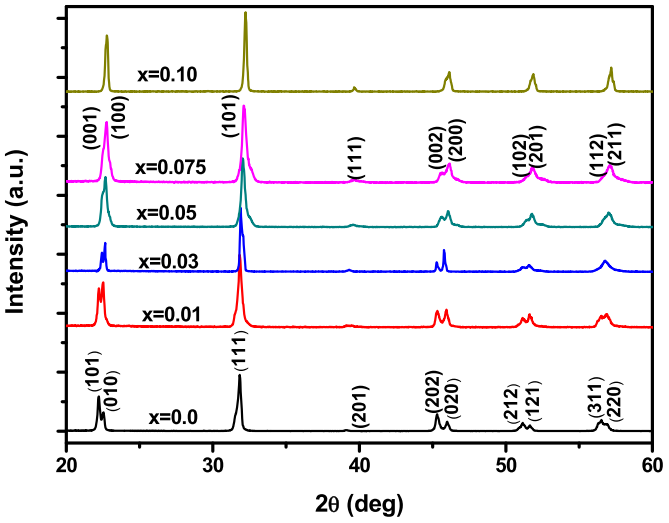

Fig. 1. XRD patterns of $(1-x)$ KNNLT $-$ xST ceramics.

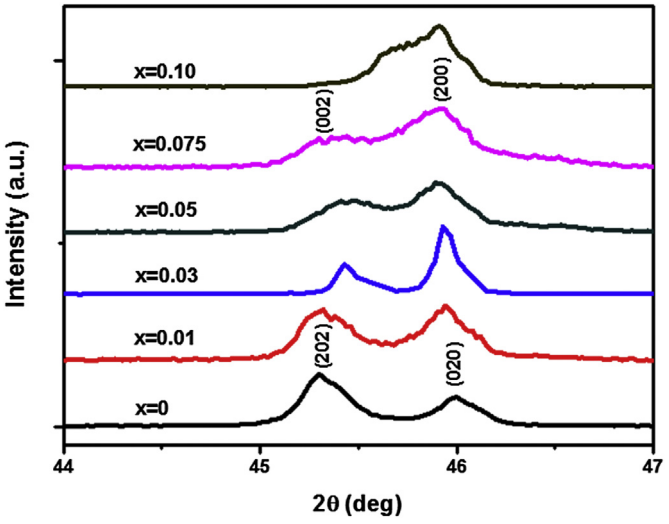

Fig. 2. Magnified XRD patterns of $(1-x)$ KNNLT $-$ xST ceramics in the range $44° < 2\theta < 47°$.

It should be noted that XRD patterns of sintered ceramics via MWS were similar to those of CFSed counterparts. Therefore in this part, only XRD patterns of CFSed ceramics are shown.

3.2. Microstructure analysis

Scanning electron microscopy images of $(1-x)$ KNNLT $-$ xST compounds which were optimally sintered via CFS and MWS are shown in Figs. 3 and 4, respectively. According to Fig. 3, CFS results