

# Piezoelectric response of $K_{0.5}Na_{0.5}NbO_3$ designed by sintering engineering

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Since the discovery of the piezoelectric properties of lead zirconate-titanate (PZT) solid solution, namely near its morphotropic phase boundary region (MPB), those materials exhibiting piezoelectricity have been largely studied, wherein PZT still stands as a prototype for electromechanical applications. However, lead based materials are poisonous in nature and thus they should be substitute for lead-free materials to assure sustainability. Though lead-based materials have higher piezoelectric response, there are other promising friendly environment compounds, namely  $K_xNa_{(1-x)}NbO_3$  [1]. For  $x=0.5$  (0.5KNN), the high-temperature cubic symmetry changes to a non-symmetric ferroelectric tetragonal structure at  $T_3=700$  K. At  $T_2=465$  K it becomes orthorhombic, stabilizing in a rhombohedral symmetry below  $T_1=135$  K [2]. Recently, theoretical calculations have predicted piezoelectric response enhancement when  $T_3$  become closer to  $T_2$ . Moreover, it has been suggested that sintering conditions can act significantly on the magnitude of the  $[T_2, T_3]$  interval. To unravel the effect of sintering conditions in 0.5KNN, ceramics were prepared by conventional sintering (CS), spark plasma sintering (SPS), and spark plasma texturing (SPT). XRD data at room conditions revealed that the two latter methods yield 0.08 and 0.16 GPa of internal stresses, respectively. It is worth emphasizing, that the emergence of these stresses have strong repercussions on the values of the piezoelectric coefficient  $d_{33}$ , increasing from 50 to 125 pC.N<sup>-1</sup> for SPS and SPT samples, respectively [3].

In this work, we present a detailed, temperature dependent, lattice dynamic study of 0.5KNN ceramics produced by the three sintering methods referred to above, using Raman spectroscopy. For the three types of sintered samples, we have observed clear critical temperature shifts, and specific different modes behaviors at  $T_1$ ,  $T_2$  and  $T_3$ . To corroborate this outcome, the temperature dependence of the polar and dielectric properties of the CS, SPS and SPT samples was studied using pyroelectric and dielectric techniques. The obtained results are discussed towards disentangling how the sintering methods tailor the piezoelectric response, in order to provide competitive lead-free materials.

[1] I. Coondoo et al., J. Advanced Dielectrics, 03, 1330002 (2013)

[2] B. Orayech et al., J. Appl. Cryst., 48, 318-333 (2015)

[3] R. Pinho et al., paper to be submitted

## Summary

## Topic

1. Multiferroics and ferroelectrics

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