

Piezoelectric properties of Spark Plasma Sintered Lead-free (K_{0.5-x}, Na_{0.5-x}, Li_{2x}) (Nb_{1-y}, Ta_y) O₃

Damien BRAULT¹, Fabien Giovannelli¹, Claire Bantignies², Bogdan Rosinski², Franck Levassort¹, Isabelle Monot-Laffez¹

¹ GREMAN, UMR 7347, Université de Tours, CNRS, INSA CVL, 37200 TOURS, France

² Vermon S. A., 37200 TOURS, France

Abstract :

Due to the toxicity of Lead and its restriction [1], piezoelectric ceramics need a substitute material as those are mostly made of Lead Zircon-Titanate (PZT) in transducers, actuators or electromechanical sensors technologies. Among the studied Lead-free family materials, Sodium Potassium Niobate (K_{0.5}Na_{0.5}NbO₃, KNN), is regarded as a promising candidate. Its substitution with Lithium, Tantalum and/or Antimony (Li, Ta et Sb), present interesting piezoelectric properties due to the existence of a morphotropic phases transition [2] enhancing its properties.

However, conditions used for conventional sintering can lead to non-stoichiometry or phases inhomogeneities due to deficiencies of alkaline [3,4]. Spark plasma sintering (SPS) can be used to overcome this problem by reducing the sintering time and temperature and reaching high 99 % densified ceramics [5,6] .

Our previous studies have shown an enhancement of the piezoelectric properties of Tantalum substituted KNN (KNN-Ta) by combining SPS and a Tantalum substitution rate between 20 and 30 % [7]. The increasement of its electromechanical properties have been explained by a better densification owing to the SPS process and the proximity of Orthorhombic-Tetragonal phase transition leading to a radial coupling factor k_p=42 %. This latter can be increased to 59 % using of a previously synthesized Nb-Ta precursor, allowing to gain a more homogeneous ceramics [8].

Such results encourage further investigations by allying a Li/Ta substitution in Spark Plasma Sintered KNN. Thus, we propose to explore different Li/Ta substitutions rate and its influence on the microstructure and piezoelectric properties. Microstructures of this Spark Plasma Sintered ceramics has been observed by SEM. Their structural changes, related to the Orthorhombic-Tetragonal transition induces by the Li/Ta rate substitution, has been investigated by XRD. And, their piezo/ferroelectric properties have been characterized by Polarization, Strain versus electric field loops.

- [1] EU - COM 2014/015, Official Journal, Off. J. Eur. Union. 57 (2014) 1–28. <http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=OJ:L:2014:015:FULL&from=EN>.
- [2] Y. Huan, X. Wang, L. Guo, L. Li, Low temperature sintering and enhanced piezoelectricity of lead-free (Na 0.52 K 0.4425 Li 0.0375)(Nb 0.86 Ta 0.06 Sb 0.08) O 3 ceramics prepared from nano-powders, J. Am. Ceram. Soc. 96 (2013) 3470–3475. <https://doi.org/10.1111/jace.12499>.
- [3] Y. Wang, D. Damjanovic, N. Klein, E. Hollenstein, N. Setter, Compositional inhomogeneity in Li- and Ta-modified (K, Na)NbO 3 ceramics, J. Am. Ceram. Soc. 90 (2007) 3485–3489. <https://doi.org/10.1111/j.1551-2916.2007.01962.x>.
- [4] L. Wu, J.L. Zhang, C.L. Wang, J.C. Li, Influence of compositional ratio K/Na on physical properties in (Kx Na1-x) NbO3 ceramics, J. Appl. Phys. 103 (2008). <https://doi.org/10.1063/1.2907866>.
- [5] R. Pinho, A. Tkach, S. Zlotnik, M.E. Costa, J. Noudem, I.M. Reaney, P.M. Vilarinho, Spark plasma texturing: A strategy to enhance the electro-mechanical properties of lead-free potassium sodium niobate ceramics, Appl. Mater. Today. 19 (2020). <https://doi.org/10.1016/j.apmt.2020.100566>.
- [6] M. Bah, F. Giovannelli, F. Schoenstein, G. Feuillard, E. Le Clezio, I. Monot-Laffez, High electromechanical performance with spark plasma sintering of undoped K0.5Na0.5NbO3 ceramics, Ceram. Int. 40 (2014) 7473–7480. <https://doi.org/10.1016/j.ceramint.2013.12.097>.
- [7] F. Jean, F. Schoenstein, M. Zaghrioui, M. Bah, P. Marchet, J. Bustillo, F. Giovannelli, I. Monot-Laffez, Composite microstructures and piezoelectric properties in tantalum substituted lead-free K0.5Na0.5Nb1-xTaxO3 ceramics, Ceram. Int. 44 (2018) 9463–9471. <https://doi.org/10.1016/J.CERAMINT.2018.02.163>.
- [8] M. Dubernet, M.J. Pitcher, M. Zaghrioui, M. Bah, J. Bustillo, F. Giovannelli, I. Monot-Laffez, Synthesis routes for enhanced piezoelectric properties in spark plasma sintered Ta-doped KNN ceramics, J. Eur. Ceram. Soc. 42 (2022) 2188–2194. <https://doi.org/10.1016/j.jeurceramsoc.2021.12.030>.