DOI 10.5162/SMSI2020/A2.5

# Electrical and Electromechanical Properties of Single Crystalline Li(Nb,Ta)$O_3$ Solid Solutions for High-Temperature Actuator Applications

*Yuriy Suhak*[1], *Bujar Jerliu*[1], *Steffen Ganschow*[2], *Dmitry Roshchupkin*[3], *Boris Red'kin*[3], *Simone Sanna*[4], *Holger Fritze*[1]
[1] *Clausthal University of Technology, Am Stollen 19B, 38640, Goslar, Germany.*
[2] *Leibniz-Institut für Kristallzüchtung, Max-Born-Str. 2, 12489, Berlin, Germany.*
[3]*Institute of Microelectronics Technology and High Purity Materials, RAS, Academician Ossipyan str. 6, 142432, Chernogolovka, Russia.*
[4]*Justus Liebig University Gießen, Heinrich-Buff-Ring 16, 35392, Gießen, Germany*
*yuriy.suhak@tu-clausthal.de*

**Summary:**
The electrical conductivity, resonance frequency and acoustic loss are determined for piezoelectric resonators which are based on Czochralski grown Li(Nb,Ta)$O_3$ solid solutions with different Nb/Ta ratios up to 700 °C. Experimental methods include impedance spectroscopy and resonance ultrasound spectroscopy. Further, the long-term behaviour of resonant properties is examined at high-temperatures. After about 400 hours at 700 °C a $LiNb_{0.5}Ta_{0.5}O_3$ resonator shows an increase in resonance frequency only by 0.1%.

**Keywords:** Piezoelectricity, high temperature, lithium niobate-tantalate, actuator, sensor.

## Background, Motivation and Objective

High-temperature stable piezoelectric actuators whose displacement can be adjusted easily by an applied voltage are required for e.g. energy conversion, aerospace or automotive industrial applications. For such materials, excellent thermal stability and large piezoelectric coefficients are required. However, common piezoelectric materials are limited by their application temperature or suffer from low piezoelectric coefficients. For example, polycrystalline ceramics show thermal instability above about 300°C [1]. Quartz type crystals from langasite ($La_3Ga_5SiO_{14}$) family are not suitable for actuators due to their low piezoelectric coefficients [2]. Lithium niobate ($LiNbO_3$, LN) and lithium tantalate ($LiTaO_3$, LT) attract substantial scientific and industrial interest because of their excellent electro-optical, piezoelectric and acoustic properties. However, their high-temperature usage is limited by thermal instability of LN and low Curie temperature of LT. Recently, attention has been attracted by Li(Nb,Ta)$O_3$ (LNT) solid solutions that combine potentially the advantages of the end members of the material system [3]. The current work explores electrical conductivity, resonant frequency and loss of LNT resonators with different Nb/Ta ratios as a function of temperature and time and in a wide oxygen partial pressure ($p_{O2}$) range.

## Specimens and Measuring Techniques

The crystals, used in this study were grown by Czochralski technique at the Institute of Microelectronics Technology and High Purity Materials, Russian Academy of Sciences, Moscow, and at the Institute for Crystal Growth, Berlin. The high-temperature experiments are performed on platinum-electroded Y-cut and Z-cut samples in a gas-tight tube furnace, which allows working temperatures up to 1000 °C. The adjustment of $p_{O2}$ is realized using an oxygen ion pump in the range from $10^{-18}$ to $10^{-3}$ bar by adjusting the oxygen content in Ar/$H_2$ gas mixtures [4].

The electrical conductivity σ is determined by impedance spectroscopy in the frequency range from 1 Hz to 1 MHz using an impedance/gain-phase analyzer (Solartron 1260). The investigations of acoustic losses are performed by means of resonant ultrasound spectroscopy on Y-cut and Z-cut LNT resonators, operated in the thickness-shear mode and in the thickness mode, respectively, using a high-speed network analyzer (Agilent E5100A). Detailed description of measuring techniques is given elsewhere [5].

## Results and Discussion

Electrical conductivity of $LiNb_{0.88}Ta_{0.12}O_3$ and $LiNb_{0.5}Ta_{0.5}O_3$ samples, measured in air in the temperature range from about 400 °C to 700 °C is shown in Fig. 1 and compared to that of pure LN and LT. As seen from the figure, the samples exhibit similar conductivity that increases linearly in the Arrhenius presentation, indicating that it is governed by a single thermally activated process in the measured temperature range. The activation energies vary between 1.2 eV and 1.3 eV for LN and LT, respectively. The observation follows the general trend of similar conduction mechanisms in LN and LT at temperatures below 700 °C [6]. Earlier, it was shown in [7] that the electrical conductivity of congruent $LiTaO_3$ shows an activation energy of 1.2 eV in the temperature range of 350–800 °C. The authors concluded that the conductivity is governed by mobile lithium vacancies. Similarly to $LiTaO_3$, our previous study shows that the lithium ion migration via lithium vacancies is the main transport mechanism in $LiNbO_3$ and the activation energy, determined for the congruent $LiNbO_3$ is equal to 1.3 eV [8].

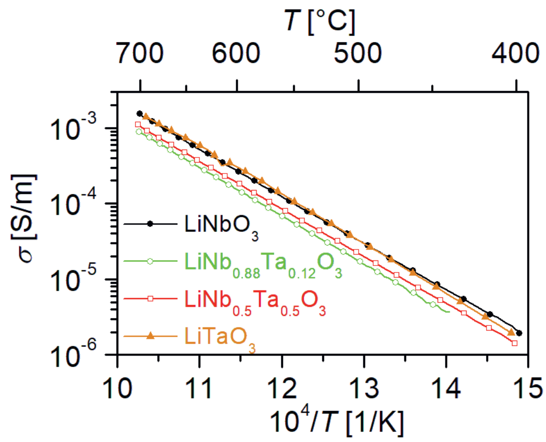


*Fig. 1. Conductivity of Li(Nb,Ta)$O_3$ samples as a function of temperature.*

Further, the measurements of electrical conductivity as a function of $p_{O2}$ revealed, that the decrease of $p_{O2}$ leads to the conductivity increase in all measured samples. However Ta-rich samples show much smaller increase, comparing to LN indicating that increased Ta content improves the stability of Li(Nb,Ta)$O_3$ at low $p_{O2}$.

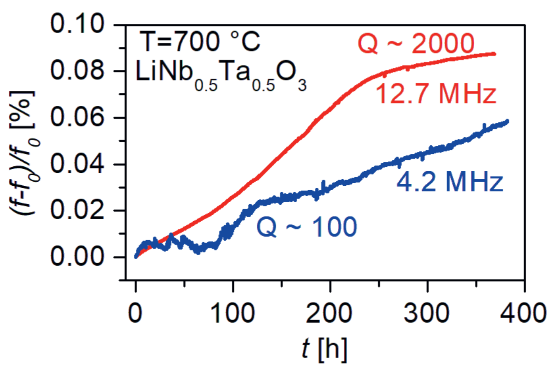


*Fig. 2. Time-dependent relative change of the frequency of a $LiNb_{0.5}Ta_{0.5}O_3$ resonator.*

Finally, in order to determine the stability of the resonant properties of LNT, the change of the resonance frequency ($f_R$) of the $LiNb_{0.5}Ta_{0.5}O_3$ specimen is examined at 700 °C in air as a function of time. Fig. 2 shows the change of the $f_R$ for the 1st and 3rd harmonics, relative to initial values measured when 700 °C was reached ($f_0$). As seen from the figure, $f_R$ steadily increases, showing however a shift of less than 0.1% only relative to $f_0$. The Q-factor equals 100 and 2000 for the 1st and the 3rd harmonics, respectively.

## Conclusions

In summary, the electrical and electromechanical properties of LNT were investigated at high temperatures and low oxygen partial pressures. The conductivity measurements reveal similar magnitudes and activation energies for all measured samples that suggests similar conduction mechanisms. At low $p_{O2}$, Ta-rich samples show improved stability compared to LN. The change of resonance frequency of the $LiNb_{0.5}Ta_{0.5}O_3$ specimen after about 400 operating hours at 700 °C in air is less than 0.1 %.

## References


[1] V. Segouin, B. Kaeswurm, K.G. Webber, L. Daniel, Temperature-dependent anhysteretic behavior of co-doped PZT, *J. Appl. Phys*. 124, 104103 (2018); doi: 10.1063/1.5040556.

[2] K. Shimamura, H. Takeda, T. Kohno, T. Fukuda, Growth and characterization of lanthanum gallium silicate $La_3Ga_5SiO_{14}$ single crystals for piezoelectric applications, *J. Cryst. Growth* 163, 388-392 (1996); doi: 10.1016/0022-0248(95)01002-5.

[3] I. G. Wood, P. Daniels, R. H. Brown, A. M. Glazer, Optical birefringence study of the ferroelectric phase transition in lithium niobate tantalate mixed crystals: $LiNb_{1-x}Ta_xO_3$, J. *Phys.: Condens. Matter* 20, 235237 (2008); doi: 10.1088/0953-8984/20/23/235237.

[4] M. Schulz, H. Fritze, C. Stenzel, Measurement and control of oxygen partial pressure at elevated temperatures, *Sensors and Actuators B* 187, 503-508 (2013); doi: 10.1016/j.snb.2013.02.115.

[5] H. Fritze, High-temperature bulk acoustic wave sensors, *Meas. Sci. Technol*., 22, 12002 (2011); doi: 10.1088/0957-0233/22/1/012002.

[6] A. Krampf, M. Imlau, Yu. Suhak, H. Fritze, S. Sanna, $LiNbO_3$: Model ferroelectric oxide or unique compound?, *In press*.

[7] A. Huanosta and A. R. West, The electrical properties of ferroelectric $LiTaO_3$ and its solid solutions, *J. Appl. Phys*. 61, 5386-5391 (1987); doi: 10.1063/1.338279.

[8] A. Weidenfelder, J. Shi, P. Fielitz, G. Borchardt, K. D. Becker, H. Fritze, Electrical and Electromechanical Properties of Stoichiometric Lithium Niobate at High-Temperatures, *Solid State Ionics* 225, 26-29 (2012); doi: 10.1016/j.ssi.2012.02.026