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Electrical and Electromechanical Properties of Single Crystalline Li(Nb,Ta)O_3 Solid Solutions up to 700 °C

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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 resonant impedance spectroscopy. Further, the long-term behaviour of resonant properties is examined at high-temperatures. After operation for about 400 hours in air 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

Piezoelectric actuators that can be operated at high-temperatures are in high demand for e.g. energy conversion, aerospace or automotive industrial applications. Such devices can generate movements in micrometer range at relatively low voltage. For such materials, excellent thermal stability and large piezoelectric coefficients are required. However, common piezoelectrics 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 possess excellent thermal stability but their piezoelectric coefficients are too low for actuating applications [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

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 Leibniz 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.

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 piezoelectric 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 [4].

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 400–700 °C is shown in Fig. 1 and compared to that of 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 increase with the Ta-content in LNT system from 1.2 eV to 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 [5]. Earlier, it was shown in [6] 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 LN is equal to 1.3 eV [7].

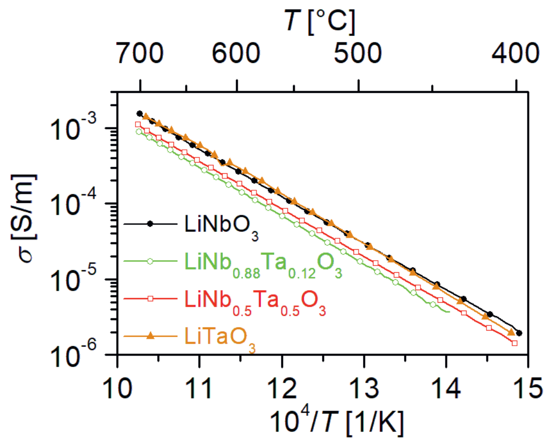

Fig. 1. Conductivity of $Li(Nb,Ta)O_3$ samples as a function of temperature.

Further, the measured acoustic losses of $LiNb_{0.88}Ta_{0.12}O_3$ and $LiNb_{0.5}Ta_{0.5}O_3$ specimens are shown in Fig. 2 in the Arrhenius plot and compared to those of pure LN, determined in [7]. As seen from Fig. 2, the losses in LNT are substantially lower in the measured range, which implies e.g. improved accuracy in frequency determination of such resonators.

Finally, in order to determine the stability of the resonant properties of LNT, the change of the resonance frequency (f_R) of the $LiNb_{0.5}Ta_{0.5}O_3$ sample is studied at 700 °C in air as a function of time during 400 h of uninterrupted thermal treatment. The measurements revealed, that f_R steadily increases with time, showing however a shift of less than 0.1% only, relative to f_0. The Q-factor at 700 °C 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. The losses in $LiNb_{0.5}Ta_{0.5}O_3$ resonators are found to be more than one order of magnitude lower, than those of congruent LN resonators. The change of resonance frequency of the $LiNb_{0.5}Ta_{0.5}O_3$ after about 400 operating hours at 700 °C in air is less than 0.1 %.

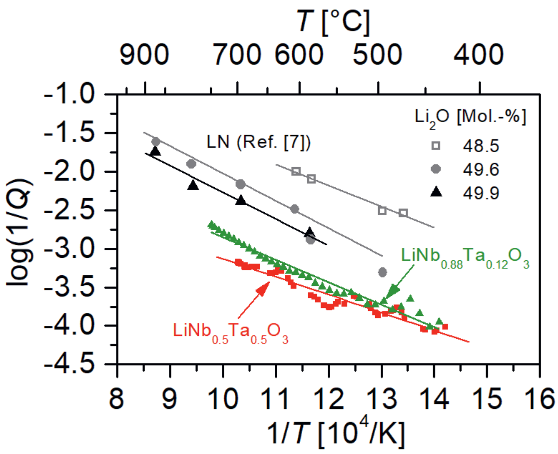

Fig. 2. Acoustic losses in $Li(Nb,Ta)O_3$ samples as a function of temperature.

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