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The frequency measurements of dielectric properties in the phase transition range in the $Li_{0.02}Na_{0.98}NbO_3$

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Measurement of the temperature, time and frequency changes of dielectric permittivity and dielectric loss in polycrystalline $Li_{0.02}Na_{0.98}NbO_3$ have been carried out. The dielectric permittivity ε has been measured in the temperature range 300K $< T <$ 750 K. The results obtained point out to the existence of polar microregions above T_c and a complicated domain structure below T_c.

Key words: *dielectric permittivity, dielectric loss, ferroelectric*

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1. Introduction

The developing progress of the present-day technique spurs the need of permanent research of newer and newer materials as well as the elaboration of their obtaining. The compounds of ABO_3-type with perovskite structure are one of the most interesting classes of materials. And among them the niobates are of great interest from the point of view of fundamental research as well as regarding their applications. The sodium niobate is the best known of all niobates [1,2], which is an antiferroelectric with very interesting dielectric and semiconducting properties [3,4]. The properties of ferro- and antiferroelectric solid solutions of lithium niobate in sodium niobate $Li_xNa_{1-x}NbO_3$ [5,6] are also very interesting.

The $NaNbO_3$ samples show the following transitions: orthorombic–543K–tetragonal–653K–cubic. Krajnik [7] investigated the phase transition of $NaNbO_3$:$LiNbO_3$-system by determining the dielectric permittivity in different fields and at different temperatures. He reported that ferroelectric pseudotetragonal and orthorombic phases emerged, when $LiNbO_3$ content increased.

The aim of the present paper was to investigate the effect of the electric field frequency on the temperature and time changes of dielectric permittivity ε and dielectric loss $\tan\delta$.

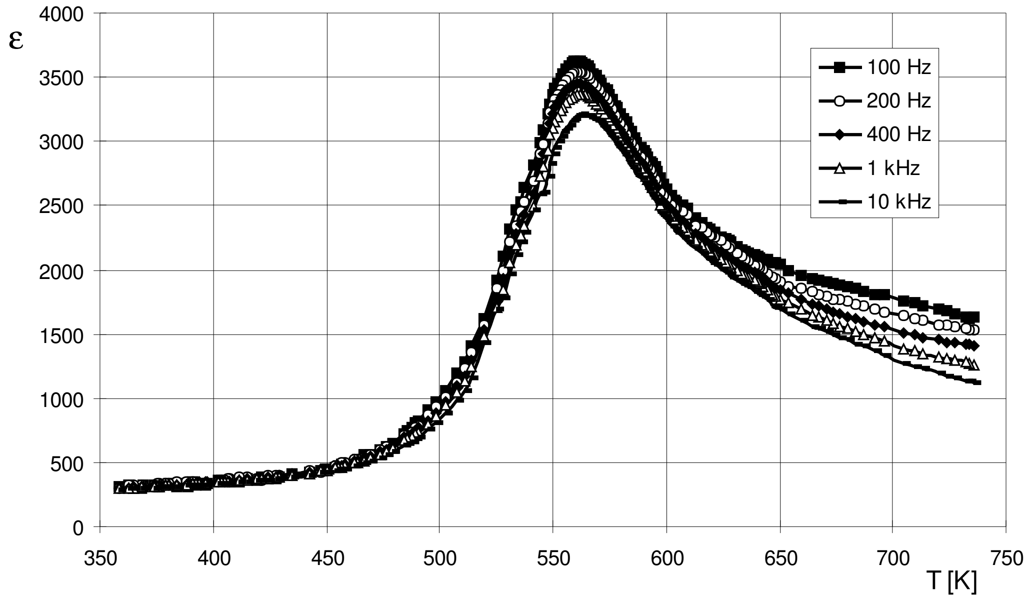

Figure 1. Temperature changes of dielectric permittivity for $Li_{0.02}Na_{0.98}NbO_3$ at cooling.

2. Experiment

The temperature, frequency and time measurements of electric permittivity were made by means of a computerized system based on BM 595 RLCG-meter at measuring electric field level 400 V/m. The measurements were performed in the temperature range 300 K–750 K and in the frequency range 100 Hz–20 kHz at a controlled rate 4 K/min on cooling. For time dependent measurements, the sample was being annealed in the temperature of 800 K during 1 hour in order to eliminate the internal and near-electrode stress. The temperature of the sample was stabilized with an accuracy of ± 0.2 K. The sample used in the experiment had the shape of a disk with the surface equal to 48 mm^2 and the thickness equal to 2.38 mm. The $Li_{0.02}Na_{0.98}NbO_3$ polycrystal has been obtained by synthesis of Na_2CO_3, Nb_2O_5 and Li_2CO_3 [6]. The sample was prepared in the Institute of Solid State Physics at the University of Latvia in Riga.

3. Results

Temperature changes of dielectric permittivity ε and dielectric loss $\tan\delta$ of the polycrystal on cooling for different frequencies of the measuring field are presented in figures 1 and 2. The maximum of dielectric permittivity is observed on cooling at about 560 K.

It can be noticed, that the maximum value of dielectric permittivity ε decreases with the increase of the frequency of measuring field. On the other hand, the temperature, at which it occurs, increases with the increase of frequency. Thermal hysteresis between heating and cooling cycles is equal to 16 K.

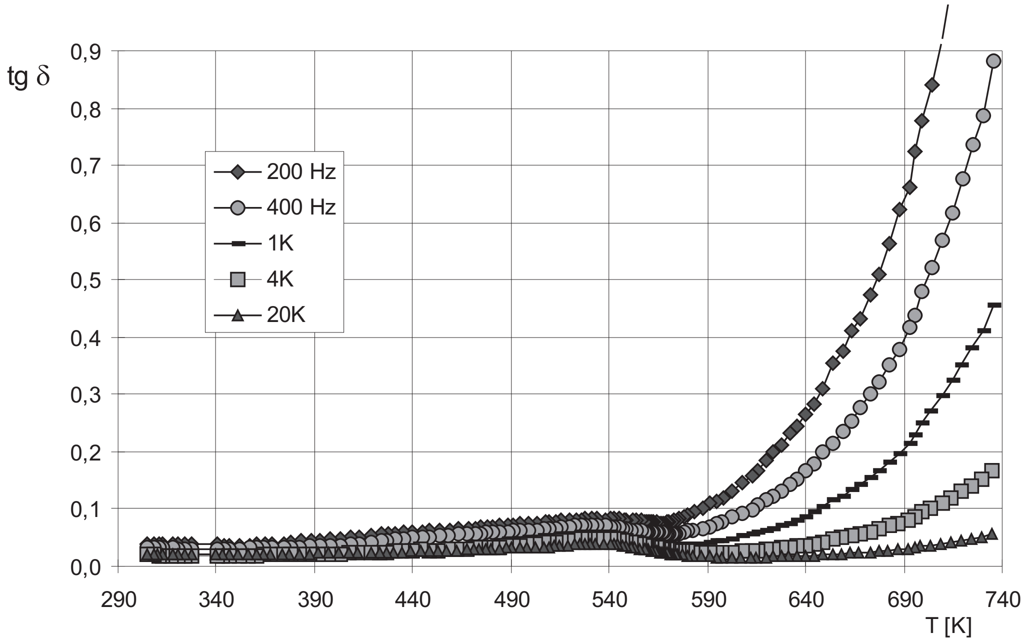

Figure 2. Temperature changes of dielectric losses for $Li_{0.02}Na_{0.98}NbO_3$ at cooling.

Analysing the temperature dependence of $\tan\delta$ we can notice the anomaly close to the temperature of 540 K (this anomaly is well seen especially for low frequencies). In the whole range of the investigated temperatures the value of $\tan\delta$ decreases with the increase of frequency.

The time changes of dielectric permittivity and dielectric loss for $Li_{0.02}Na_{0.98}NbO_3$ for several temperatures (below and above phase transition) and frequencies are shown in figures 3 and 4. Strong time dependence of dielectric permittivity in temperature region of 500–580 K was observed. The time changes of ε (as a dependence $\varepsilon/\varepsilon_{(t=0)}$ on t) are presented in figure 3. These changes depend both on temperature and frequency. In the range of phase transition the time changes $\varepsilon(t)$ are the largest. Time changes of dielectric loss $\tan\delta$ (as a dependence $\tan\delta/\tan\delta_{(t=0)}$ on t) are presented in figure 4. For $\tan\delta$ the time changes increase with the increase of frequency and their values also depend on temperature.

The investigations carried out show that the dielectric permittivity ε and $\tan\delta$ change in time according to the logarithmic law [8].

4. Summary

Based on the investigations performed the following conclusions can be drawn:

- the substitution of sodium ions by lithium ions leads to the arising of polar regions above T_c [9],
- the time changes of dielectric permittivity ε below T_c can be connected with the changes of domain structure [8,10], tending to achieve the stable state,

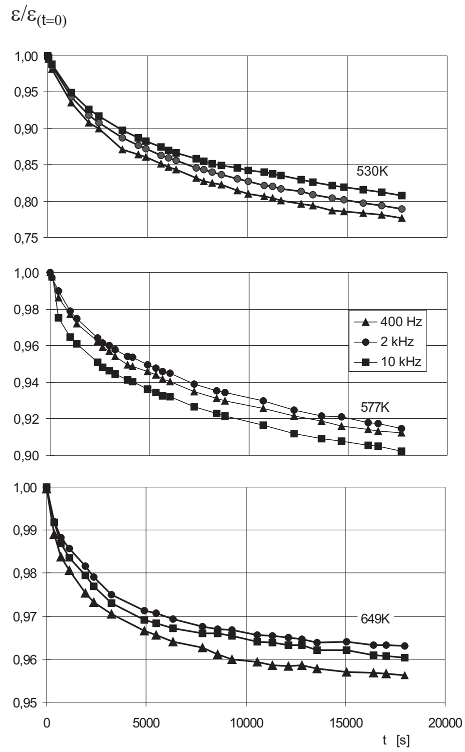

Figure 3. Time changes of dielectric permittivity for $Li_{0.02}Na_{0.98}NbO_3$.

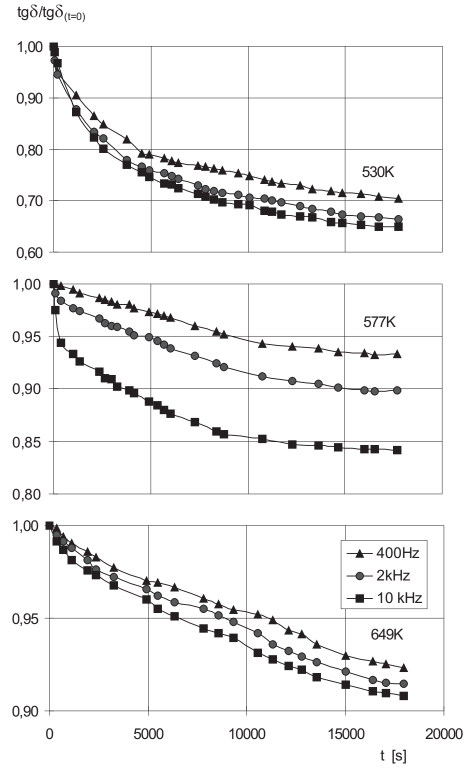

Figure 4. Time changes of dielectric losses for $Li_{0.02}Na_{0.98}NbO_3$.

characterized by the smallest value of free energy, in the material,

- the defects of the lattice and the impurities strongly effect the ε value and its changes versus the temperature and versus the frequency of the applied electric field.

Taking into account the latter conclusion the following explanation can be proposed. While annealing the ceramic and depositing electrodes some defects arise. After cooling some of them are frozen. The most active point defects, which are oxygen vacancies, play the basic role in solid solution $Li_xNa_{1-x}NbO_3$ and in $NaNbO_3$. These defects, frozen in cooling process, become the centres of capture for the electron emitted from the metallic electrodes to the inside of the sample. These centres, which are being polarized under the effect of the alternating field, give rise to polar microregions. They contribute to the entire polarization of the sample and cause the increase of dielectric permittivity value in the cooling process. The interaction of these microregions leads to the emerging of time dependent dielectric permittivity as well as to its dependence on the frequency of the measuring field.

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Частотні вимірювання діелектричних властивостей $\mathrm{Li_{0.02}Na_{0.98}NbO_3}$ в області фазового переходу

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Проведено вимірювання температурних, часових і частотних залежностей діелектричної проникності і діелектричних втрат у полікристалічному $\mathrm{Li_{0.02}Na_{0.98}NbO_3}$ в області температур 300K $< T <$ 750 K. Отримані результати вказують на існування полярних мікроскопічних областей вище T_c та складної доменної структури нижче T_c.

Ключові слова: *діелектрична проникність, діелектричні втрати, сегнетоелектрик*

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