Condensed Matter Physics, 1999, Vol. 2, No. 4(20), pp. 727–730

CONDENSED MATTER PHYSICS

Investigation of dielectric properties of $\mathrm{Li_xNa_{1-x}NbO_3}$ ceramics for $\mathrm{x} = 1$ and $\mathrm{x} = 4$

W.Bąk, B.Garbarz, W.Śmiga, C.Kuś

Institute of Physics and Computer Science, Pedagogical University,
2 Podchorążych Str., 30-084 Kraków, Poland

Received September 1, 1998

The a.c. electrical response of $\mathrm{Li}_x\mathrm{Na}_{1-x}\mathrm{NbO}_3$ (LNN) solid solution in low frequency range (100 Hz–20 kHz) has been analysed as a function of temperature (300 K–750 K). A complex picture of the dependence of the studied properties on the chemical composition has been obtained. The data indicate the presence of relaxation and transport processes as well as their thermally activated character.

Key words: *$\mathrm{Li}_x\mathrm{Na}_{1-x}\mathrm{NbO}_3$, ceramics, dielectric properties*

PACS: *77.84.Dy, 77.22.-d*

1. Introduction

Dependence of dielectric properties of $\mathrm{Li}_x\mathrm{Na}_{1-x}\mathrm{NbO}_3$ (LNN) solid solution on its chemical composition is well known. In particular, phase transformation, conductivity and dielectric permittivity of LNN have been studied [1–3]. However, some discrepancies in the results are observed, and more systematic studies should be undertaken to establish relations of the properties to the composition of the solid solution.

The system $\mathrm{NaNbO_3}$–$\mathrm{LiNbO_3}$ is of interest, especially as Li content in LNN increases. These materials would be good candidates for the fabrication of piezoelectric and piroelectric components for high temperature purposes.

The investigated polycrystalline sample was obtained using the conventional ceramic technology. The application of a.c. measurements method enables us to observe the polarization processes and to gain the information about the charge transport mechanism.

2. Experimental

The ceramic samples of LNN used in our measurements had the shape of disks of 8–9 mm diameter and 2–3 mm thickness. Samples with sputter deposited *Pt*-electrodes were used in our measurements. All the experiments have been performed under normal atmospheric conditions. The temperature of the samples, in the range between 300 K and 800 K, was measured by means of a chromel-alumel thermocouple with 0.5 K accuracy. The dielectric measurements were performed by means of RLC meter (model BM 595) at different fixed frequencies between 100 Hz and 20 kHz. In all measurements the RLC meter was set to a parallel mode.

3. Results and discussion

The real part of the dielectric permittivity ε as a function of the temperature for two measurement frequencies (1kHz and 10kHz) is shown in figure 1a for Li-content x=0.01 and in figure 1b for Li-content x=0.04. As the content of Li increases ε generally increases.

The thermal hysteresis of permittivity for two *Li*-contents when measurements are made at the increasing and the decreasing temperature is due to the observed thermal hysteresis of the dielectric anomaly caused by the antiferroelectric-paraelectric (A-P) phase transition of $NaNbO_3$ [4]. There is a small shift (cooling process) of the temperature of the ε-maximum to lower temperatures with the increasing Li-content.

The behaviour of $\tan\delta$ as a function of the temperature changes with the frequency, as it is shown in figure 2a for Li-content x=0.01 and in figure 2b for Li-content x=0.04. For low frequency (<1kHz) there is a fast increase of $\tan\delta$ above 600 K. In the whole range of the investigated temperatures the value of $\tan\delta$ decreases with the increase of frequency.

Figures 3a,b show the conductivity σ versus temperature for different measurement frequencies and different Li-contents ((3a) x=0.01 and (3b) x=0.04). For Li-content of x=0.01 there is a minimum at T=400 K, which tends to disappear as the

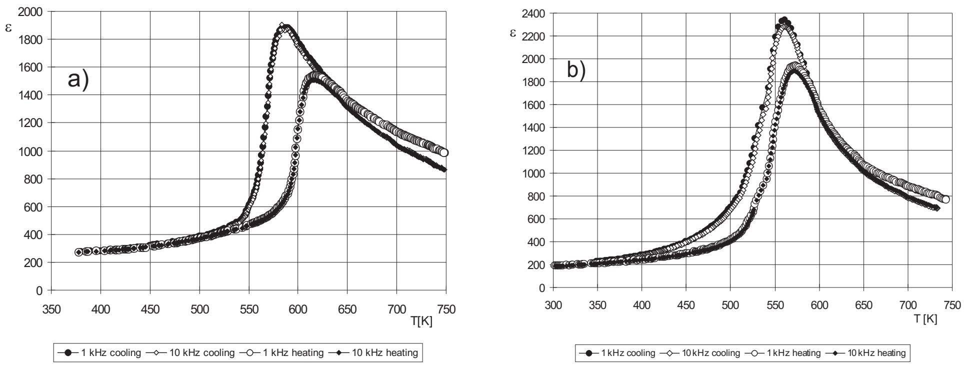

Figure 1. Real value of the permittivity ε as a function of the temperature at two measurement frequencies for: (a) LNN, x=0.01; (b) LNN, x=0.04.

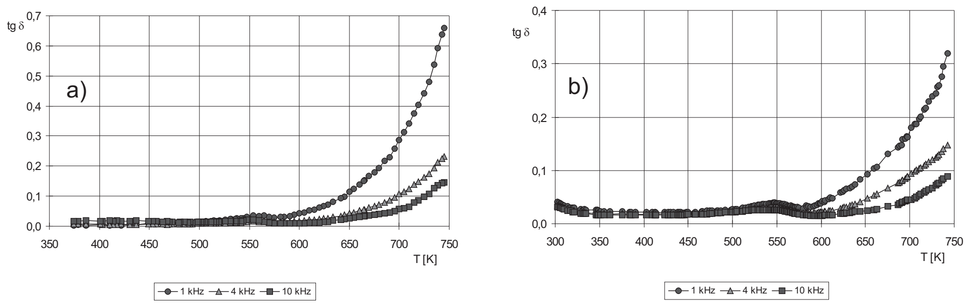

Figure 2. Temperature dependencies of $\tan\delta$ at several measurement frequencies for: (a) LNN, x=0.01; (b) LNN, x=0.04.

frequency and Li-content increases. The low frequency (<1kHz) and low temperature (<400 K) change in the temperature coefficient of the conductivity with the appearance of a minimum (figure 3a) was also found in lithium [5] and sodium [6] niobates and sodium tantalate [7] at the similar temperature and lower frequencies (1 mHz to 1 Hz). The minimum in conductivity could be related to the possible existence of conduction mechanism of small polarons (i.e., localized charge carriers permitted by a certain degree of structural disorder or polarizability of the surroundings of the charge carriers [8]) at low temperature. At low temperatures the short range tunnelling gives place to conductivity values corresponding to long range hopping, with higher activation energy that would correspond to higher temperatures due to a low mobility of these localized charge carriers. The minimum in the thermal behaviour of ε is, therefore, explained as the result of the change in the conductivity mechanism, from tunnelling to hopping, from short range to long range mechanisms, at this temperature [6].

References

1. Kuś C., Ptak W.S., Śmiga W., Bąk W. Temperature dependence of dielectric properties of nonstoichiometric $NaNbO_3$. // Acta Universitatis Wratislaviensis, 1988, No. 1084, p. 169-175.
2. Jankowska I., Krzywanek K., Kuś C. The investigation of metastable states in $Na_{1-x}Li_xNbO_3$ polycrystals near the diffused phase transition. // Ferroelectrics, 1992, vol. 127, No 4, p. 83-88.

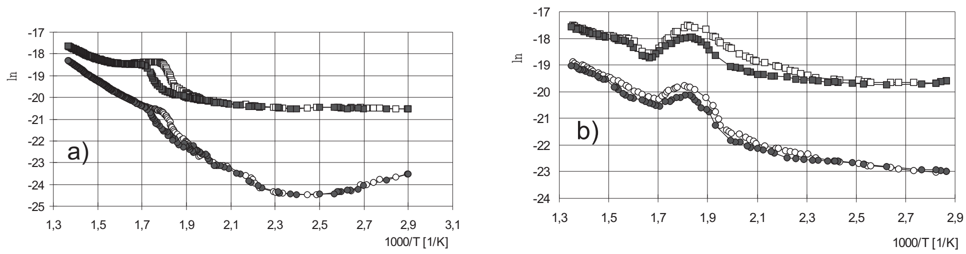

Figure 3. Conductivity σ as a function of the temperature at two measurement frequencies for: (a) LNN, x=0.01; (b) LNN, x=0.04.

3. Pardo L. et al. Temperature behaviour of structural, dielectric and piezoelectric properties of sol-gel processed ceramics of the system $LiNbO_3$-$NaNbO_3$. // J. Phys. Chem. Solids, 1997, vol. 58, No 9, p. 1335-1339.
4. Kuś C., Bąk W., Ptak W.S., Śmiga W. Antiferroelectric- paraelectric phase transformation in nonstoichiometric $NaNbO_3$. // Ferroelectrics, 1988, vol. 81, p. 277-280.
5. Bąk W., Kuś C., Ptak W.S., Śmiga W. Very low frequency study on transport and relaxation phenomena in $LiNbO_3$ single crystal. // Ferroelectrics, 1992, vol. 126, p. 179-184.
6. Bąk W., Kuś C., Ptak W.S. The transport properties of polycrystalline $NaNbO_3$ determined from immitance spectra at very low frequencies. // Ferroelectrics, 1991, vol. 115, p. 105-111.
7. Aleksandrowicz A., Wójcik K. Electrical properties of single crystals and ceramic samples of $NaTaO_3$. // Ferroelectrics, 1989, vol. 99, p. 105-113.
8. Jonscher A.K. Dielectric Relaxation. London, Chelsea Dielectric Press, 1993.

Дослідження діелектричних властивостей $Li_xNa_{1-x}NbO_3$ керамік у випадках $x = 1$ і $x = 4$

В.Бонк, Б.Гарбаж, В.Сьміга, Ч.Кусь

Інститут фізики та обчислювальної техніки,
Педагогічний університет,
Польща, 30-084 Краків, вул. Подхоронжих, 1

Отримано 1 вересня 1998 р.

Досліджено електричний відгук в змінному полі твердих розчинів $Li_xNa_{1-x}NbO_3$ (LNN) як функцію температури (300 K–750 K) в низькочастотній області (100 Hz–20 kHz). Отримано комплексну картину залежності досліджених величин від хімічного складу. Отримані дані свідчать про наявність релаксаційних і транспортних процесів та про тепловий характер їх активації.

Ключові слова: *$Li_xNa_{1-x}NbO_3$, кераміки, діелектричні влатстивості*

PACS: *77.84.Dy, 77.22.-d*