The Electric Properties of $Na_{0.5}Bi_{0.5}TiO_3$ and $0.87Na_{0.5}Bi_{0.5}TiO_3$ - $0.13BaTiO_3$ Single Crystals

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The sodium bismuth titanate $Na_{0.5}Bi_{0.5}TiO_3$ (NBT) and $Na_{0.5}Bi_{0.5}TiO_3$-based solid solutions are considered as promising ecologically friendly materials for piezoelectric devices and electromechanical transducers [1]. In the abstract we study the effect of heat treatment on the electric properties of NBT and $0.87Na_{0.5}Bi_{0.5}TiO_3$- $0.13BaTiO_3$ (0.87NBT-0.13BT) single crystals. The objective of the study is to reveal the contributions of intrinsic defects to the electric properties of the crystals studied.

The permittivity ε and conductivity σ were measured by using an AC bridge P5083 in the frequency range 0,5÷100 kHz and the temperatures diapason 300K÷800 K. The as-grown samples were annealed in air at the temperature 1100K (1 h) and then in vacuum at 1100K (2 h). Temperature- frequency dependencies of ε and σ were measured for as-grown samples and after each heat treatment. The spectra of complex impedance were measured by Tesla BM-507 impedance meter operating in frequency range $5Hz\div5\cdot10^5Hz$. Temperature of the samples was regulated within the interval from 600K to 900 K.

It was shown that ε and σ for the studied crystals strongly depended on heat treatment atmosphere and temperature. Annealing at T_{ann}=1100 K in air resulted in disappearance of ε relaxation peak near 670 K and significantly decreased conductivity. Subsequent annealing at T_{ann} in vacuum restored low-frequency relaxation maximum of ε but practically did not change σ. The data obtained are discussed in assumption that heat treating in air decreases content of oxygen vacancies V_O, whereas annealing in vacuum generates additional amount of V_O [2].

In order to clarify the mechanisms of charge transport in NBT and 0.87NBT-0.13BT crystals we studied the spectra of complex impedance $Z^*(\omega)=Z' - i\cdot Z''$. Before the measurements the samples were annealed in air at 900K for 1h in order to remove low-frequency dispersion of ε near 670 K. Experimental spectra were plotted as diagrams on the complex (Z'-Z'') plane and discussed on the basis of the equivalent circuits approach. In the studied temperature-frequency range the hodographs of NBT crystals consist of two arcs which are described by impedance of two parallel RC circuits which are connected in series. The hodographs of 0.87NBT-0.13BT crystals consist of a single arc. It is supposed that charge transfer in the bulk of NBT contains the contributions of electronic and ionic conductance. Ionic conductance can be result of oxygen vacancies $V_O^{\bullet\bullet}$ movement. Electrons can hop via traps such as F^+ centers. The low-frequency arc in the experimental hodographs reflects electron conductivity in the near-electrode regions. The effect of $BaTiO_3$ additive on the mechanisms of charge transfer in NBT crystals is discussed.

[1] S. Nahm, S. Priya, *Lead-Free Piezoelectrics*, Springer, New York, 2012.
[2] T.V. Kruzina, V.M. Sidak, M.P. Trubitsyn et al., *Ferroelectrics* **462** (2014) 140.