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Dielectric properties of polycrystalline $(Ba_{0.40}Sr_{0.60})O_3$

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ABSTRACT

Purpose: Purpose of this paper is to qualify of the degree and the causes of broadening of the phase transition in the polycrystalline solid solution $(Ba_{0.40}Sr_{0.60})TiO_3$ (BS60T).

Design/methodology/approach: Polycrystalline samples of $(Ba_{0.40}Sr_{0.60})TiO_3$ (BS60T) were prepared by calcinations method at temperature 1570K. Images of the morphology of the samples were taken by means of electron microscope Philips SEM 525M at room temperature. Dielectric measurements were performed with application of Quatro Cryosystem 4.0 Agilent Precision LRC meter HP4824A equipped with WinDETA 5.62 software Novocontrol. Measurements were taken under cooling with 2K/min speed. Measuring electric field frequency was from the range 20 Hz - 1 MHz.

Findings: The dielectrometry was applied to measure complex dielectric permittivity and other dielectric functions of ferroelectric (BS60T). It was affirmed, that 60% substitution of Sr ions changed the type of phase transition. Weak dependence of temperature T_m=208 K (temperature of maximum electric permittivity ε') on frequency of the external electric field testifies about diffused character of the phase transition (DPT). The polar character of this material was also observed in the paraelectric phase (to 360 K). This observation is connected with the occurrence of polar clusters in this phase.

Research limitations/implications: Results can be used in order to describe the phase transition modifications in the solid solutions with ferroactive substitutions in sublattice B of the perovskite.

Originality/value: Value of this work relies on the experimental examination of the electric properties of BS60T solid solution. The low value of phase angle in the paraelectric phase is connected with the occurrence of the polar regions (clusters).

Keywords: Ferroelectric; Solid solution; Phase transition; Dielectric properties; Cluster

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PROPERTIES

1. Introduction

Barium strontium titanate $(Ba_{0,40}Sr_{0,60})TiO_3$ (BS60T) is the solid solution of the ferroelectric $BaTiO_3$ (BT) and the ferroelectric $SrTiO_3$ (ST) [1, 2]. The dielectric behaviour of $(Ba_{1-x}Sr_x)TiO_3$ (BSxT) changes from typical for ferroelectrics (BT) to typical for ferroelectrics with diffused phase transition (DPT-[3, 4]) with increasing Sr substitution. Barium Titanate shows 3 structural phase transitions: cubic–tetragonal–orthorhombic–rhombohedral (C-T-O-R). The first is para-ferroelectric (PF) phase transition (PT) (ca. 400K). ST shows PF PT in temperature ca. 105 K. This material in region from 140 K to 470 K is paraelectric. Increase of Sr substitution in $(Ba_{1-x}Sr_x)TiO_3$ leads to decrease of temperature of PF PT and maximum value of real part of the dielectric permittivity ε‘ [5]. The studied solid solution (BS60T) do not show, in area of the PF PT, an essential dependence of the T_m on frequency of the external electric field. The possibility of changes of physical properties was cause of investigations of the polycrystalline BS60T samples. These investigations were based on methods of dielectric spectroscopy and electron microscopy SEM.

These materials are used as ceramics to building thermistor, condensers and piezoelectric elements in actuators [6, 7].

2. Experimental

The polycrystalline BS60T-sample was obtained by the calcinations method (1570 K). The structure BTS-10 was investigated with use of the electron microscope Philips SEM 525M at room temperature.

A sample was diameter 10mm and thickness 1.5 mm it was covered by silver electrodes. The dielectric measurements were performed by automatic device (QUATROKRIO 4.0 with analyzer Agilent type 4824A and BDS 1100). The measurements were in cooling, with speed 5 K/min, in range of frequency from 20 Hz to 1 MHz executed.

3. Results and discussion

Figure 1 presents the structure of polycrystalline BS60T obtained by SEM method at room temperature. Clear crystallites are visible on Figure 1. Their sizes have carried out since several to a dozen or so micrometers.

Figure 2 presents a value of real part of the dielectric permittivity as temperature function ε‘(T) for polycrystalline BT sample. In this figure three structural phase transition are visible: cubic–tetragonal–orthorhombic–rhombohedral (C-T-O-R).

Figure 3 shows a value of real part of the dielectric permittivity as temperature function ε‘(T) for polycrystalline BS60T sample. One can see a diffused maximum of ε' at temperature 208 K. This temperature is independent on frequency.

The 60% concentration of the paraelectric $SrTiO_3$ leads to strong decrease of the value ε_m and the temperature T_m. The change of character of the PF PT from sharp PT in pure BT to diffused phase transition (DPT) in BS60T was observed.

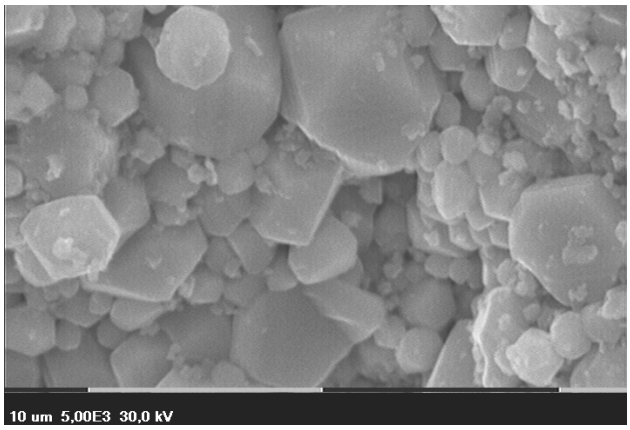

Fig. 1. The crystalline structure of BS60T ceramic obtained by electron microscope (magnification 5 x10^3)

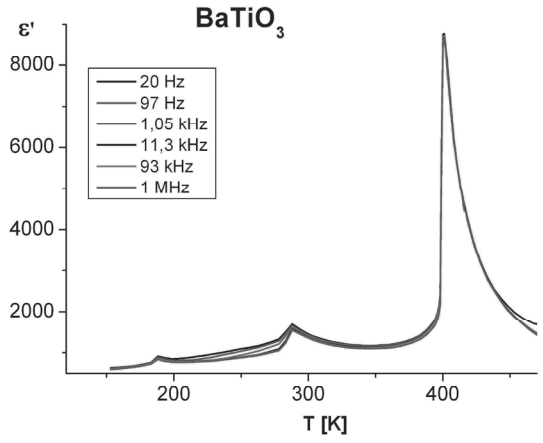

Fig. 2. Dependence of ε’(T) for polycrystalline BT

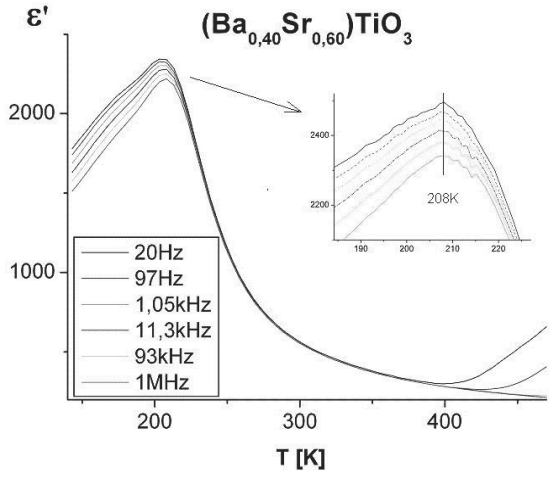

Fig. 3. Dependence of ε’(T) for polycrystalline $Ba_{0,40}Sr_{0,60}TiO_3$

The following formula describes ferroelectric materials with DPT:

$$\varepsilon^{-1} = \varepsilon_m^{-1} + A(T - T_m)^{\gamma} \quad (1)$$

where ε_m is the maximum value of the electric permittivity ε', T_m is the temperature of maximum of the electric permittivity ε', A and γ are constants.

In DPT the value of γ is close to 2. These values were compiled in Figures 4-5, where:

$$y = \varepsilon^{-1} - \varepsilon_m^{-1}, \quad (2)$$

$$x = T - T_m. \quad (3)$$

These figures present dependence (logy) on (logx) for frequencies 20 Hz and 1 MHz. The value of γ parameter (γ = 1.64) testifies to DPT. Smaller values of this parameter in higher temperatures and the higher frequency (1MHz) suggest the behaviour of the weakly diffused phase transition. Similar anomaly was observed in solid solution BS30T.

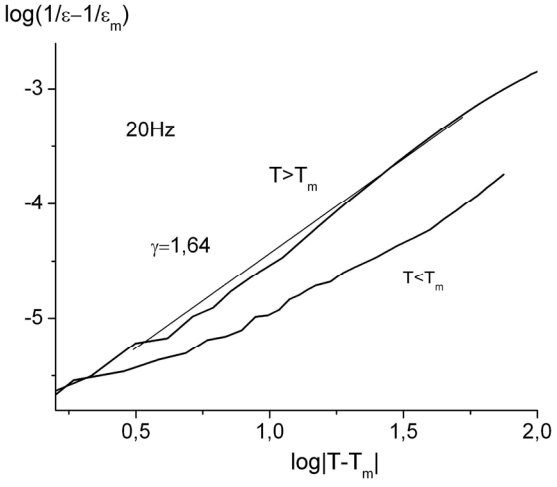

Fig. 4. Dependence of (logy) on (logx) for polycrystalline BS60T (20 Hz)

Fig.6 show dependencies of the phase angle Φ(T) as a function of temperature for polycrystalline $Ba_{0,40}Sr_{0,60}TiO_3$. Observed (at 360 K) value of phase angle, approximate to -90deg, testifies to polar character of material in the paraelectric phase too. The polar regions (clusters) are cause of this state. The clusters are created in high temperatures [8, 9]. Increase of dipolar polarization P_d is connected with growth of the polar clusters [10-12]. Their evolution (increase and interaction) leads to PF PT. The lack of maximum of Φ near temperature 208 K testifies that total, dipolar polarization in this area does not change.

The changes of clusters configuration leads to their liability and flexibility on the applied electric field. The violent increase of value of the phase angle in higher temperatures, which starts for low frequencies, testifies to freeing the charges [13-15]. The freed (by breaking up clusters) of charges leads to change of conductivity type to electron one.

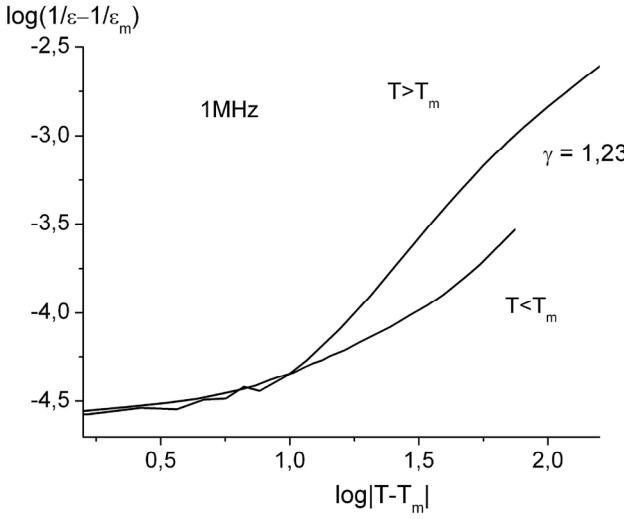

Fig. 5. Dependence of (logy) on (logx) for polycrystalline BS60T (1 MHz)

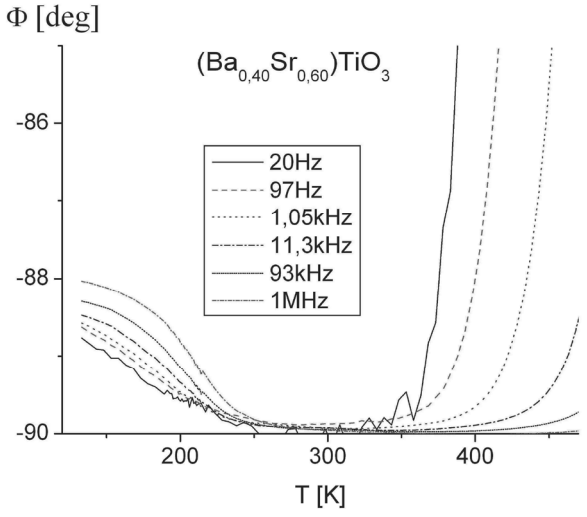

Fig. 6. Dependence of phase angle Φ(T) for polycrystalline $Ba_{0,40}Sr_{0,60}TiO_3$

Figure 7 presents electric conductivity Sig' as the temperature function for BS60T sample. Increase of frequency about 5 orders of size leads, in low temperatures, to approximately the same increase of the electric conductivity. In this figure it is also visible, that by cooling near T_m an increase of the electric conductivity about two orders of size take place. It confirms the earlier suggestion connected with the changes of system of clusters in region of the PF PT.

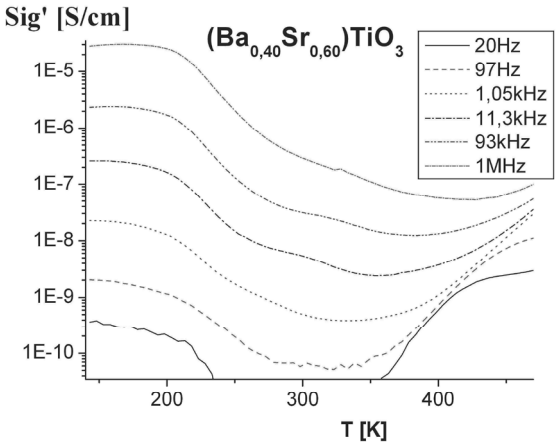

Fig. 7. The dependence Sig'(T) for polycrystalline $BSr_{0.60}T$

4. Conclusions

The results of investigations of the dielectric properties for polycrystalline BS60T were presented. The occurrence of the DPT was affirmed. This fact testifies that even big Sr concentration does not lead to relaxor or glassy behaviour [16, 17]. The similarity of Ba and Sr ions is cause of this behaviour. The transition of this type was observed in solid solutions BSxT for lower concentrations of strontium also as well as in different solid solutions $Ba(Ti_{0,90}Sn_{0,10})O_3$ as well as $Ba(Ti_{0,90}Zr_{0,10})O_3$. Below 360K the value of phase angle is approximate equal to -90^0. This suggests occurrence of the polar regions (clusters) in the whole region of temperatures. Their evolution leads to large liability of system of clusters. Possibility of modification of physical properties, the temperatures PT and their character can be crucial for future applications.

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