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Relaxor phase transition of polycrystalline $(Ba_{0.90}Sr_{0.10})(Ti_{0.75}Sn_{0.25})O_3$

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Properties

ABSTRACT

Purpose: The purpose of this work was to determinate the influence of the nonferroactive 25% Sn substitutions in (sublattice B) on changes of the character of phase transition (PT) in comparison with pure barium titanate $BaTiO_3$ (BT).
Design/methodology/approach: The dielectrometry method was applied in order to measure complex dielectric permittivity and other dielectric functions of ferroelectric $(Ba_{0.90}Sr_{0.10})(Ti_{0.75}Sn_{0.25})O_3$ (BSr10TSn25).
Findings: It was affirmed, that 25% substitution of Sn ions changed the type of phase transition. The transition was strongly diffused and typical for relaxor materials. The strong dependence of temperature T_m (peak of the electric permittivity ε') on frequency of the applied electric field was observed. It means, that this material belongs to relaxor type. The polar character of this solution was also observed in a broad temperature range (in the paraelectric phase too). This behaviour is connected with the occurrence of polar regions (clusters).
Practical implications: The results can be used in order to describe changes of PT in the relaxor solid solutions with nonferroactive substitutions in sublattice B.
Originality/value: Value of this work relies on the experimental examination of the dielectric properties of (BSr10TSn25) solid solution. The temperature of the glassy transition T_f and the activation energy was calculated. The low value of the phase angle is connected with the existence of the polar regions. The dielectric phase transition is connected with change of interactions, of average range, in the system of clusters.
Keywords: Ferroelectric; Solid solution; Phase transition; Dielectric properties; Cluster

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1. Introduction

The solid solution $(Ba_{0,90}Sr_{0,10})(Ti_{0,75}Sn_{0,25})O_3$ is one of the ferroelectric materials of type (A',A")(B'B")O_3. Both components BaTiO3 (BT) and $SrTiO_3$ (ST) of this solution are ferroelectrics. The pure barium titanate (BT) forms four structural phases [1-3].. At high temperatures (in the paraelectric phase) it has a cubic structure. Three phase transitions take place during decrease of temperature: first to tetragonal phase T (~ 400 K), second to rhombic phase, O (~ 300 K) and third to rhombohedral phase, R (~210 K). The strontium titanate shows the phase transition at temperature 105K. Tin is not ferroactive and therefore it shift the temperature of phase transition in the direction of lower temperature [2]. Increase of tin concentration causes the growth of maximum value of the dielectric permittivity ε'_{max}. Further increase of the concentration of tin reduces the ε'_{max} value.

The aim of the presented work is to determine the influence of nonferroelectric Sn on the physical properties of BSr10T and also to analyze the change of a character of phase transition in this material. The ions Ti and Sn have the similar parameters of external orbitals. Statistical layout B' and B" atoms can cause broadening of phase transition temperature region [4, 5]. This fact can have essential meaning in applications. The analysis of influence of the nonferroactive ions on structure of crystalline perovskite can explain changes of physical properties of ferroelectric materials.

Barium titanate is used as capacitor ceramics, piezoelectric transducers, thermistor and chemical sensors [6-8].

2. Experimental

The polycrystalline samples of BSr10TSn0 and BSr10TSn25 were obtained by the calcinations method in temperature 1620 K. The dielectric measurements were executed by automatic device (QUATRO KRIO 4.0 with LCR Agilent 4824A meter and BDS 1100 cryostat). The materials were investigated under cooling conditions and within frequency range from 20 Hz to 1 MHz.

3. Results and discussion

Figure 1 present the real component of the electric permittivity ε'(T) vs. temperature for BS10T and BSr10TSn25. the temperature dependence of the electric permittivity ε'(T) shows three clear maximums for BSr10T. They involve following phase transitions: C (cubic) – T (tetragonal) – O (orthorhombic) – R (rhombohedral). The function ε'(T) shows a broad phase transition for BSr10TSn25.

Figure 2 present peaks of phase transitions related to frequency of applied electric field for BSr10TSn25 sample. This fact is typical for relaxor materials.

From the glass model one can write down the following equation involving $T_{m,v}$ values for different frequencies:

$$\nu = \nu_0 \exp\left(\frac{E_a}{k_b\left(T_{m,\nu} - T_f\right)}\right) \qquad (1)$$

where: ν and ν_0 are the relaxation frequency at temperature T and temperature high enough, k_b is the Boltzman constant, E_a is the activation energy of polar region and T_f is the freezing temperature of polar regions.

Figure. 3 shows the (lnν) as a function of $(1000/(T_{m,v}-T_f))$. From this dependence the glass temperature T_f = ca. 120 K was obtained as well as the activation energy E_a equal ca. 0,5 eV.

Figure 4 shows, the low values of the phase angle Φ ot temperatures below 400K. This means, that in this area BSr10TSn25 behaves as a polar dielectric. The observed phase transitions in surroundings of the T_m temperatures, should have the connection with changes of structure of polar areas (that are brought into being in high temperatures). Below 450 K the values of phase angle Φ belongs to the interval from – 90deg to –80deg. The changes of phase angle with temperature means that the polar substructures play decisive role in transition process.

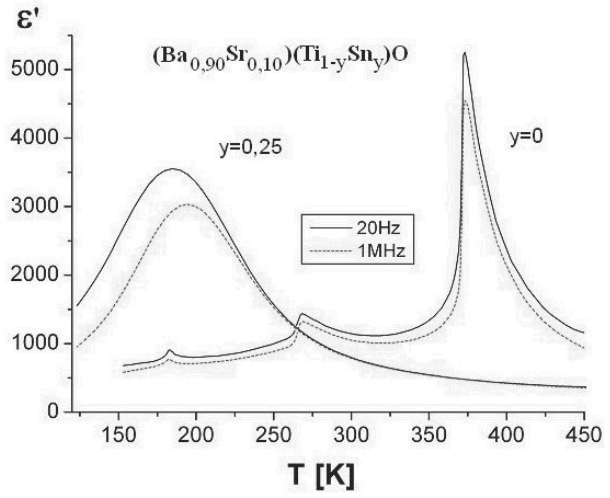

Fig. 1. Dielectric constant ε' as a function of temperature for polycrystalline BS10T and BSr10TSn25 samples

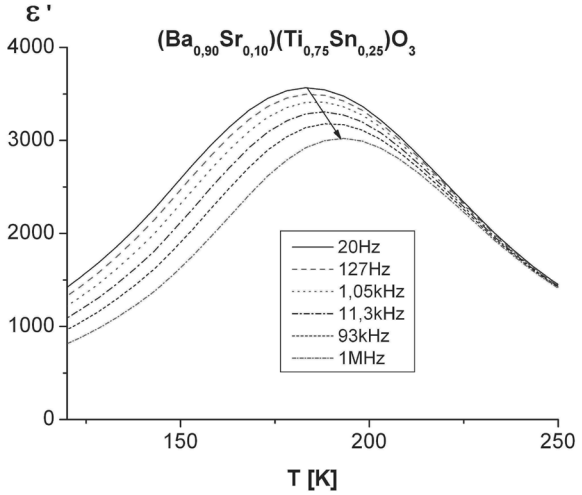

Fig. 2. Dielectric constant ε' as a function of temperature for polycrystalline BSr10TSn25 sample (peak)

They create clusters change with temperature. These clusters are the source of dipolar polarization P_d [9-11]. This polarization originates in short range interactions of electric dipoles. The growth of clusters leads to creation their long range structural and electric collectivization. This is connected with change of density of domain walls too [12, 13].

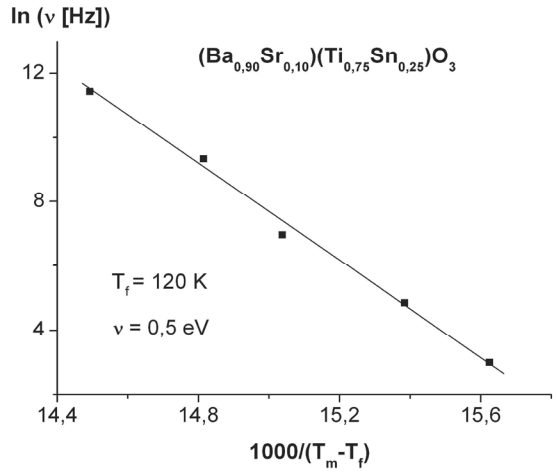

Fig. 3. The dependence (lnν) on (1000/(T_m-T_f)) for polycrystalline BSr10TSn25 sample

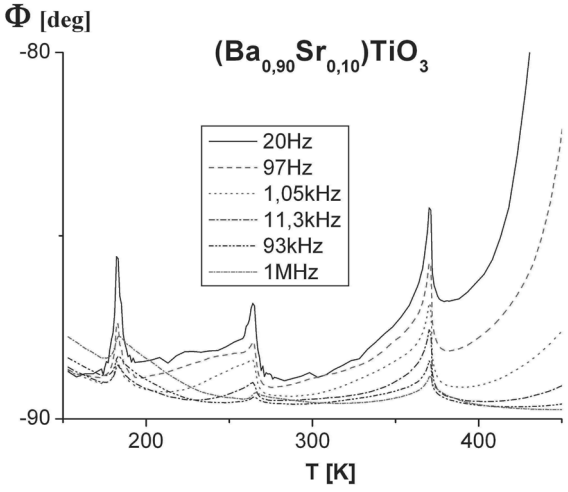

Fig. 4. Phase angle Φ as a function of temperature for polycrystalline BS10T sample

Figure 5 shows the value of phase angle Φ as a function of temperature for polycrystalline BSr10TSn25 sample. In this figure a disappearing of local maxim of phase angle is visible. One can also observe small growth values of Φ in temperatures below 200K.

4. Conclusions

The study presents results of dielectric measurements of the BSr10TSn25. It was confirmed that the 25% Sn-substitution of the nonferroactive ions Sn (in sublattice B) lead to strong diffuseness and to lowering of the temperatures of paraelectric – ferroelectric PT. This PT has a relaxor character. Sn-ions lead to freezing (T_f) of cubic structure below the temperature (T_m) of the paraelectric – ferroelectric PT. This behavior is similar to $Pb(Cd_{1/3}Nb_{2/3})O_3$ [14] and $Ba(Ti_{0,70}Sn_{0.30})O_3$ [15]. In the last case, nonferroactive ions Sn make impossible transition from cubic to tetragonal structure.

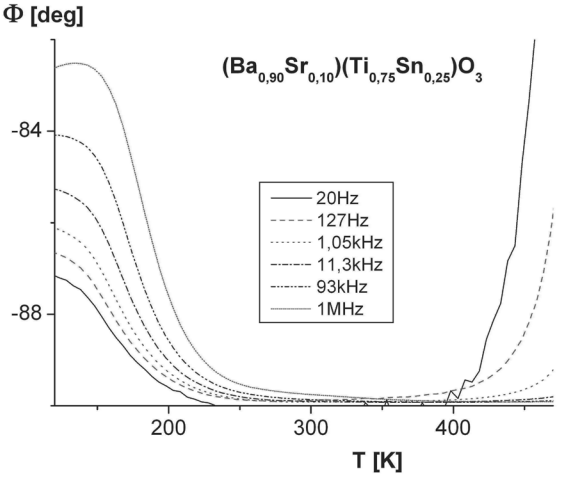

Fig. 5. Phase angle Φ as a function of temperature for polycrystalline BSr10TSn25 sample

The change in concentration of the nonferroactive Sn ions can be applied in practice to change of work temperature for electromechanical elements. Polycrystalline BSr10TSn25 can be used as capacitor and piezoelectric materials.

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