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Phase formation and relaxor properties of lead-free perovskite ceramics on the base of sodium-bismuth titanate

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Lead-free relaxor oxides on the base of rhombohedral perovskite sodium-bismuth titanate $(Na_{0.5}Bi_{0.5})TiO_3$ (NBT) are being intensively studied in order to replace widely used Pb-based ones [1, 2]. Advantage of the NBT-based compositions comprises .presence of polar nanoclusters leading to high mobility of boundaries “domain walls/polar clusters” [3].

We studied influence of A-cations substitutions on structure parameters, microstructure, dielectric, and relaxor ferroelectric properties of solid solutions in the systems $(Na_{0.5}Bi_{0.5})_{1-x}B_xTiO_3$ with $B = K^+$, La^{3+}, x = 0-0.1 [4]. Ceramic samples were prepared by the two-step solid-state reaction method at temperatures T_1 = 973 K (6 h), and T_2 = 1200-1400 K (2 h).

The samples were characterized using the X-ray Diffraction, Scanning Electron Microscopy, Second Harmonic Generation (SHG), and Dielectric Spectroscopy methods. To improve density of ceramics overstoichiometric KCl and LiF additives were used.

The pseudocubic unit cell parameter increased in modified ceramics in accordance with the ionic radii changes. Main size of grains decreased with x increasing in both systems.

Ferroelectric phase transitions were revealed at as steps near ~ 400 K and as peaks at ~ 600 K in the dielectric permittivity versus temperature curves. Phase transitions near ~ 400 K demonstrate marked relaxor behavior typical for the NBT-based compositions conditioned by the presence of polar nanoregions in a nonpolar matrix [3]. Using the SHG method increase in the spontaneous polarization value was observed in ceramics with B = K with x increasing. While decrease in SHG signal in ceramics with $B = La^{3+}$ points to enhancement of relaxor properties due to possible decrease in the size of polar nanoregions. In the samples modified by KCl additional anomalies in the $\varepsilon(T)$ dependences were revealed at T > 900 K above the ferroelectric phase transition due to formation of vacancies in the oxygen sublattice.

At the room temperature, non monotonous changes of the dielectric parameters were observed in modified compositions studied confirming prospects of new lead-free materials development by modification of NBT-based compositions with aliovalent cation substitutions.

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