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Dielectric and ferroelectric properties of Nb-doped $Ba_{0.8}Sr_{0.2}TiO_3$ ceramics

L. Yang · W. Q. Cao

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Abstract Ferroelectric and dielectric properties were investigated for $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$ ceramics with different Nb_2O_5 concentrations. The relations between the ceramic structures and those properties were discussed. The $Ba_{0.8}Sr_{0.2}TiO_3$ doping with 0.01mol% Nb_2O_5 appears to have a strong ferroelectric effect and better dielectric properties. The max permittivity (ε_{max}) is up to 7,521.3 and $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$ ceramics has higher permittivity even at room temperature. The permittivity presents broadened curves at large temperature ranges, which suggests a non Curie–Weiss behavior near the transition temperature. The diffuse phase transition coefficient (δ) for $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$ doping with 0.01mol% Nb_2O_5 reaches 0.098, and its *P–E* loop expresses a diffusing curve. The remanent polarization ($2P_r$) and coercive field are 31.3 μC/cm^2 and 10 kV/cm, respectively. The *P–E* loop presents a diffusing curve, which is relative to the relaxor characteristic.

Keywords Ferroelectric ceramics · Dielectric · Diffuse phase transition · $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$

1 Introduction

Barium strontium titanate, $Ba_xSr_{1-x}TiO_3$ (BST), has excellent ferroelectric and dielectric properties, which has attracted much attention. It was widely used in devices such as capacitors, phase shifting in antenna and radar equipment, ferroelectric memory for computers, etc. [1, 2]. Doping of $Ba_xSr_{1-x}TiO_3$ to fine tune its properties for particular applications is therefore an interesting research task. Doping of the $Ba_xSr_{1-x}TiO_3$ (x=0.6 and 0.7) compositions with different elements are being widely investigated [3–7] due to its high dielectric permittivity, small dielectric loss, and transition temperatures near the room temperature of the undoped system. Regarding the rise of the devices' working temperature, the present work is dedicated to the study of stabilities in high dielectric constant, ferroelectric properties, and relation between ferroelectric properties and the structure around the room temperature for the Nb-doped $Ba_{0.8}Sr_{0.2}TiO_3$ ceramics.

L. Yang · W. Q. Cao (✉)
School of Physics and Electronic Engineering, Hubei University, Wuhan 430062, China
e-mail: caowang62@yahoo.com.cn

2 Experiments

Due to the similar electronegativity and the ionic radii for both Nb^{5+} and Ti^{4+}, the compositions of the samples studied are given by $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$ (BST20), where x=0.01, 0.05, and 0.1. Ceramic samples were prepared by the conventional solid-state reaction technique using materials of $BaCO_3$, $SrCO_3$, TiO_2, and Nb_2O_5. The raw materials were calcined at 1100 °C for 2 h. The calcined mixture was ball-milled and pressed into pellet disks (10 mm in diameter, 1 mm in thickness).The samples were sintered at 1280 °C for 4 h. The crystal phase composition was studied by X-ray diffraction (XRD). The PHILIPS PW-1710 with CuKα anode (λ=1.5406 Å) was used for XRD. The 10-kV JEOL microscope model M5400 was used for analyzing the microstructure of the samples. The HP4294 was used to test the dielectric properties. The hysteresis meter (RT-6000HVS) was used to measure the *P–E* hysteresis loop.

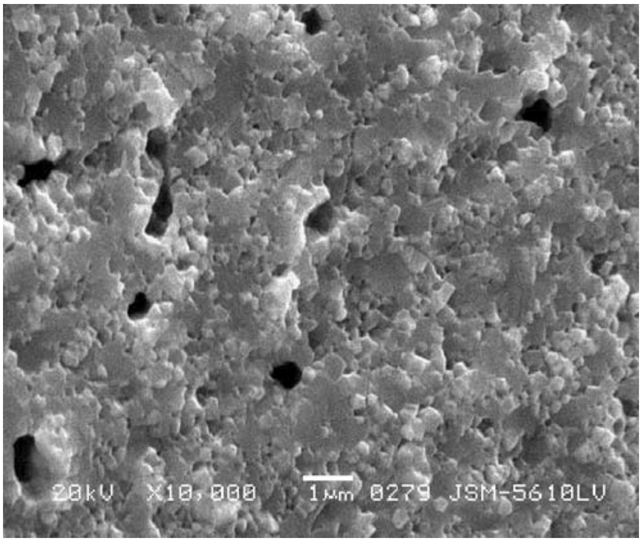

Fig. 1 The SEM of BST20 doping with 0.1mol% Nb

3 Results and discussion

Figure 1 shows the SEM micrographs of the $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$ (x=0.01). The crystal grains grow well and connected with each other tightly, and little air vents can also be seen in the ceramics.

Figure 2 exhibits the XRD patterns of the $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$ (x=0, 0.01, 0.05, and 0.1). Compared with the standard XRD pattern of the $Ba_{0.7}Sr_{0.3}TiO_3$, the samples has perfect perovskite phase. The crystal parameters grow a litter large with the increase of Nb concentration (Table 1), which can be considered as a result from the substitution of larger Nb^{5+} for small Ti^{4+}.

The dependences of the dielectric constant and the dielectric loss on temperature measured at 1 kHz for all the studied compositions are shown in Figs. 3 and 4,

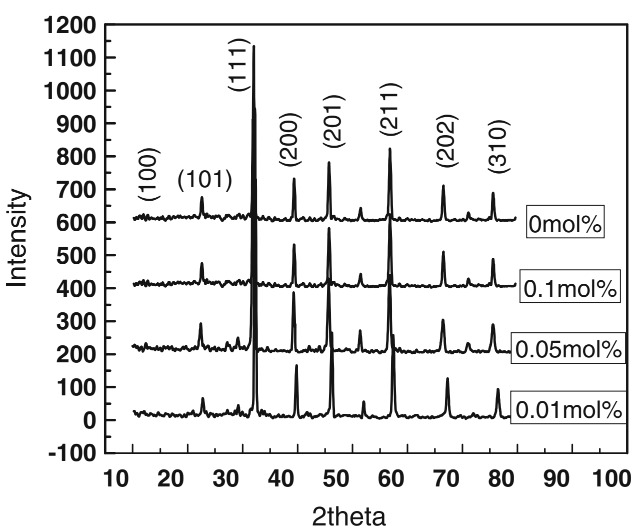

Fig. 2 XRD pattern of BST20 ceramics system

Table 1 Lattice parameters for BST20 of all composition.

Composition	a or b (Å)	c (Å)	V (Å3)
0	3.980	3.981	63.06063
0.01	3.982	3.983	63.15574
0.05	3.993	3.9998	63.77301
0.1	4.038	4.042	65.9066

respectively. The dielectric constant curve of BST20 ceramic system doping with 0.01mol% Nb presents a broadened peak, which suggest a non Curie–Weiss behavior near the transition temperature.

Figure 5 shows an explicit frequency dispersive relation of the dielectric constant. The phase transition temperature is 56 °C. There are little differences among curves at lower temperature part of the peaks, but larger differences at higher temperature part of the peaks. Therefore, the frequency dispersive property can be described with a diffuse phase transition relation, i.e., the diffuse phase transition coefficient (δ) determined by the developed Smolenskii's relation [3, 8]:

$$\frac{1}{\varepsilon}-\frac{1}{\varepsilon_{\max}}=C(T-T_{\mathrm{m}})^{\gamma} \tag{1}$$

where

$$C=\frac{1}{2\,\varepsilon_{\max}}\delta^{2} \tag{2}$$

Obviously, the transformed experiment results demonstrate a linear relation with the transformed temperature in Fig. 6. So a fitting relation of $\left(\frac{1}{\varepsilon}-\frac{1}{\varepsilon_{\max}}\right)$ to $\log(T-T_{\mathrm{m}})$ is accordant with the experiments shown in Fig. 6 with the following expression:

$$\log\left(\frac{1}{\varepsilon}-\frac{1}{\varepsilon_{\max}}\right)=-6.2+1.6\log(T-T_{\max}) \tag{3}$$

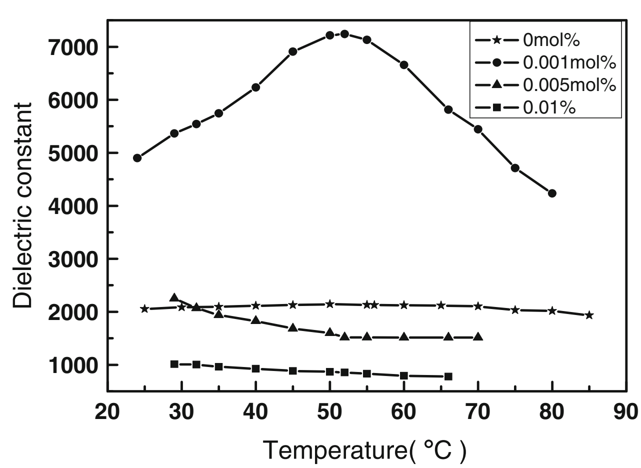

Fig. 3 Dependence of the dielectric constant with temperature

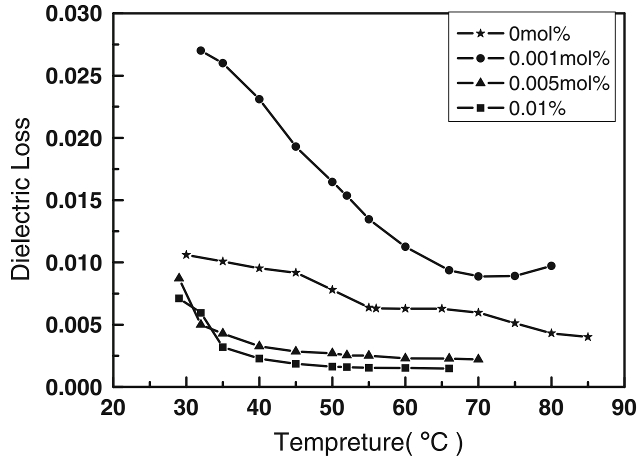

Fig. 4 Dependence of the dielectric loss with temperature

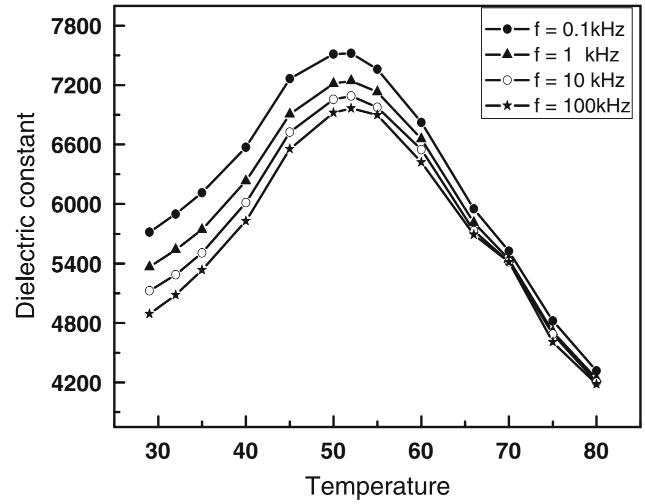

Fig. 6 Dependence of $\log(1/\varepsilon - 1/\varepsilon_{\max})$ with the $\log(T - T_{\max})$ for the BST20 ceramic system doping with 0.01mol% Nb. *Square points* are experiment results and the *line* is the Smolenskii's relation

The data in Eq. 2 means that the diffuse phase transition coefficient (δ) is 0.098, and the factor γ is 1.6.

Figure 7 shows the *P–E* hysteresis loop of the BST20 at room temperature. Figure 7a shows that the $2P_r$ is 31.3 $\mu C/cm^2$ and the E_c is 10 kV/cm for BST doping with 0.01mol% Nb. The P_r and E_c increased with the drive voltage. When the voltage increased, the polarization does not grow to the saturated line as usual, but increasing and then decreasing. There were many order and disorder microregions in the ceramics, and some regions' polaron have frozen. When the voltage increased, these polarons are stimulated and result in the polarization–voltage line diffusion such as the *P–E* hysteresis loop showed in Fig. 7(a). Figure 7(b) shows the *P–E* hysteresis loop in a different composition. It can be observed that the BST20 ceramic system doping with 0.01mol% Nb has the best ferroelectric properties.

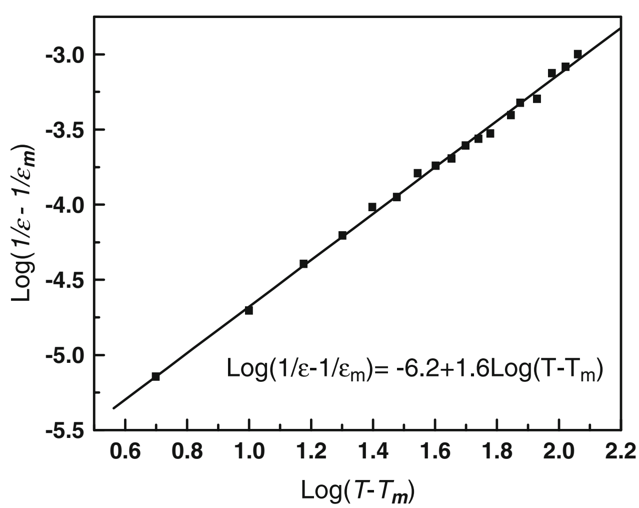

Fig. 5 Dependence of the dielectric constant with temperature for x=0.01 at different frequency

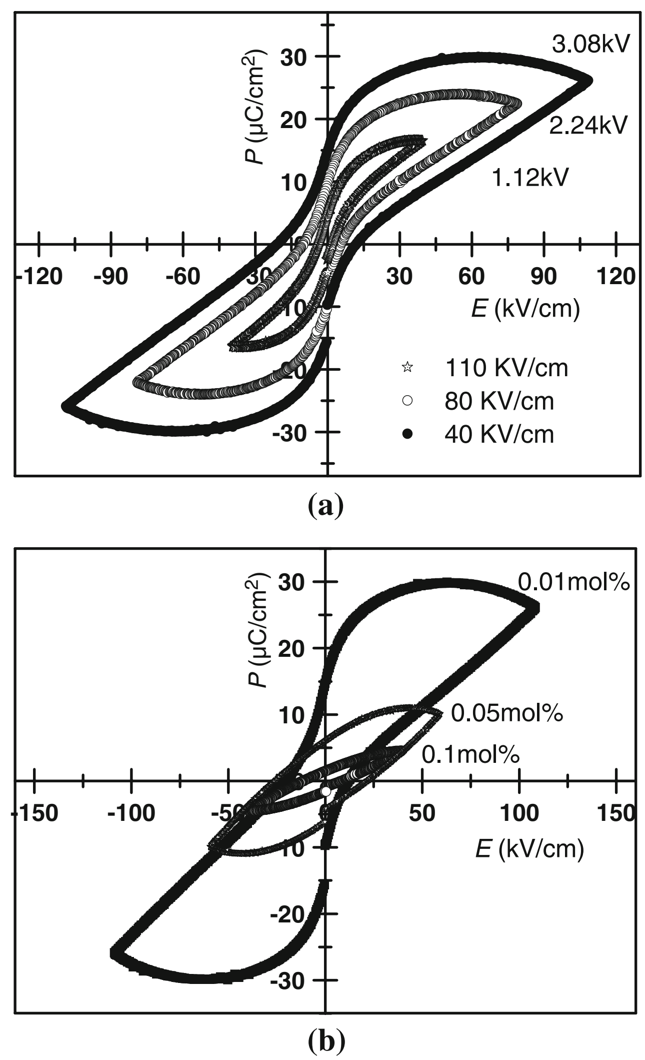

Fig. 7 (**a**) *P–E* hysteresis loop of a different voltage of BST20 doping with 0.01mol% Nb. (**b**) *P–E* hysteresis loop of different composition

4 Conclusion

The $Ba_{0.8}Sr_{0.2}Ti_{(1-5/4x)}Nb_xO_3$ ($\underline{x}$=0.01, 0.05, and 0.1) system ceramic was sintered at 1280 °C for 4 h. The $Ba_{0.8}Sr_{0.2}TiO_3$ doping with 0.01mol% Nb_2O_5 has strong ferroelectric and good dielectric properties. The max permittivity (ε_{max}) is 7,521.3 and the permittivity at room temperature remains at high value. The relation of permittivity with temperature for $Ba_{0.8}Sr_{0.2}TiO_3$ doping with 0.01mol% Nb_2O_5 presents broadened curves, which suggest a non Curie–Weiss behavior near the transition temperature near the room temperature, but follows the developed Smolenskii's relation, $\frac{1}{\varepsilon} - \frac{1}{\varepsilon_{max}} = C(T - T_m)^\gamma$, where C is $10^{-6.2}$ and γ is 1.6. The diffuse phase transition coefficient (δ) of BST system doping with 0.01mol% Nb_2O_5 was 0.098. The BST20 system ceramic performs an explicit frequency dispersive relation or a relaxor characteristic. The remanent polarization ($2P_r$) and coercive field are 31.3 μC/cm^2 and 10 kV/cm, respectively, for $Ba_{0.8}Sr_{0.2}TiO_3$ doping with 0.01mol% Nb_2O_5. And the P–E loop presents a diffusing curve, which is relative to the relaxor characteristic.

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