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EFFECT OF POLING CONDITION ON THE PIEZOELECTRIC PROPERTIES OF $0.935(Bi_{0.5} Na_{0.5})TiO_3$ - $0.065BaTiO_3$ CERAMICS

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Abstract

In this study, the effect of poling conditions on the piezoelectric properties of $0.935(Bi_{0.5} Na_{0.5}) TiO_3$ - $0.065BaTiO_3$ (BNT6.5BT) lead free ceramics was examined. The piezoelectric properties like piezoelectric constant (d_{33}) and electromechanical factors (K_p ,K_t) depend on poling field and poling temperature, while no remarkable effect of the poling time on the piezoelectric properties was detected in the range of 5-30 min.

Key Words: ceramics, poling conditions, piezoelectric properties.

Résumé

Dans cette étude, l'effet des conditions de polarisation sur les propriétés piézoélectriques de céramiques exemptes de plomb $0.935(Bi_{0.5} Na_{0.5}) TiO_3$ -$0.065BaTiO_3$ (BNT6.5BT) a été examiné. Les propriétés piezoelectriques tels que la constante piezœlectrique (d_{33}) et les coefficients de couplage électromécanique (K_p ,K_t) dépendent du champ électrique de polarisation et de la température de polarisation tandis qu'aucun effet remarquable du temps de polarisation sur les propriétés piézoélectriques n'a été détecté dans l'intervalle de 5-30 min.

Mots clefs : céramiques, conditions de polarisation, propriétés piézoélectriques.

1. INTRODUCTION

Lead zirconate titanate abbreviated as PZT ceramics have been widely used in piezoelectric applications because of their excellent piezoelectric and electrical properties [1]-[2]. However, because of the toxicity of lead oxide, the use of these ceramics has caused serious environmental problems [3]. Therefore, there is a great need to develop lead-free piezoelectric ceramics with good piezoelectric properties for replacing the lead-containing ceramics in various applications.

Bismuth sodium titanate $(Bi_{0.5} Na_{0.5})TiO_3$ (abbreviated as BNT) is considered to be an excellent candidate of lead-free piezoelectric ceramics [4]. The BNT ceramic exhibits a large remnant polarization P_r = 38 μC/cm^2, a high Curie temperature T_c = 320 °C and a phase transition point from ferroelectric to antiferroelectric T_d = 200 °C. However the use of BNT in piezoelectric application is limited by the difficulty to pole this ceramic due to its large coercive field (73 kV/cm).To improve piezoelectric and dielectric properties of BNT ceramics, various BNT-based solid solutions have been developed [5]-[6]-[7]-[8]-[9]. Among these solid solutions, $(1-x)(Bi_{0.5} Na_{0.5})TiO_3$ - $xBaTiO_3$ (BNT-*x*BT) system has been attracted a great deal of attention owing to the existence of a rhombohedral-tetragonal morphotropic phase boundary (MPB) near *x*=0.06-0.07 [10]. Compared with pure BNT, the BNT-*x*BT ceramics reveal substantially improved poling and piezoelectric properties near the MPB.

A poling operation is a requisite process to yield desired piezoelectric performances of BNT-based solid compositions. Therefore it is interest to investigate the influence of the poling condition on the piezoelectric properties of theses compositions. In this paper, the influence of the poling condition on the piezoelectric properties of $0.935(Bi_{0.5}\ Na_{0.5})TiO_3 - 0.065BaTiO_3$ (BNT6.5BT) lead-free ceramics was studied.

2. EXPERIMENTAL PROCEDURE

A conventional powder solid-state reaction method was used to prepare the samples: weighted powder of Na_2CO_3, $BaCO_3$, Bi_2O_3 and TiO_2 (at least of 99% purity) were ball-milled by planetary milling in ethanol for1 h. After calcining at 825^0C for 4 h in air atmosphere, the powder was ball-milled again and then isostatically pressed. The compacted samples were sintered at 1160^0C for 4 h in air atmosphere. The as-prepared samples were cut in disks shape of 12mm in diameter and 1mm in thickness.

The specimens were polished and electroded with a silver paste and after were poled in a silicone oil bath under different poling conditions (poling field, poling temperature and poling time) for piezoelectric measurements. The piezoelectric coefficient d_{33} was measured using a piezoelectric d_{33}-meter (Priestest PM 200) at a frequency of 100 Hz. The electromechanical coupling factors k_p and k_t were measured by the resonance and anti-resonance technique using an impedance analyzer (HP 4194A).

3. RESULTS AND DISCUSSION

3.1-Phase structure and Microstructure:

Figure.1 shows the X-ray diffraction (XRD) patterns of BNT-6.5BT ceramics sintered at 1160^0C in the 2θ range of $20\text{-}70^0$ determined by means of X-ray diffractometer (RIGAKU Miniflex) using Cu Kα radiation. From Fig.1, it can be seen that the sample displays a pure perovskite structure phase. It can be clearly seen that the (003),(021) reflections of rhombohedral phase and (200),(002) reflections of tetragonal phase appear near $39{,}84^0$ and $46{,}51^0$ respectively. This result shows that the sample exhibit co-existence of a rhombohedral- tetragonal phase.

Fig. 2 shows the SEM micrograph of BNT-6.5BT ceramics observed by with scanning electron microscope (SEM, HITACHI,S-3500N). It can be seen that the ceramics were densified. The microstructures consist of fine and homogeneous grains with an average grain size close to 1µm. Some slight porosity is observed and consists of small intergranular pores. The density was measured by Archimedes' method and was about 97.6%.

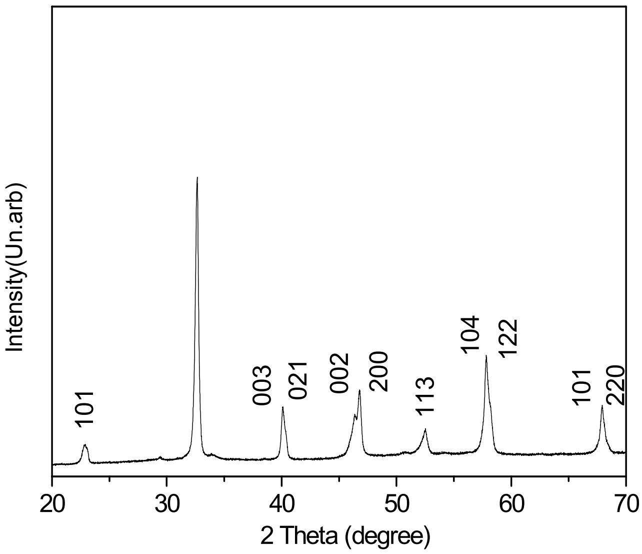

***Fig.1**: XRD patterns of $0.935(Bi_{0.5}\ Na_{0.5})TiO_3 - 0.065BaTiO_3$ ceramics sintered at 1160^0C.*

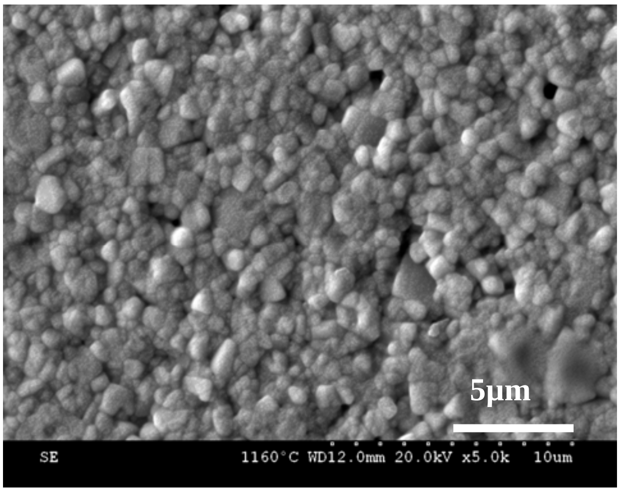

***Fig.2**: SEM micrograph of BNT-6.5BT ceramics sintered at 1160^0C.*

3.1- Effects of poling conditions on piezoelectric properties:

For revealing the influence of condition poling on the piezoelectric properties of the ceramics, we have start by poling the ceramics under poling field in the range of 30-50 KV/cm at temperature poling of 60^0C for 15 min. It was found that the poling field affected the piezoelectric properties. The piezoelectric constant d_{33} increases monotonously with poling field. An increase of poling field to 55 KV/cm did not enhance the piezoelectric properties. Moreover the application of the

poling field over 55 KV/cm leads to electrical break-down of some specimens. Thus, the poling field of 50 KV/cm was found to be the optimal field for the specimens.
In second time we have fixed a poling field at 50 kV/cm and the poling temperature ranges from 25 to 100°C for 10mn.The dependency of piezoelectric constant d_{33} and electromechanical factors (k_p and k_t) according the poling temperature for the ceramics are shown in figs. 3 and 4 respectively.

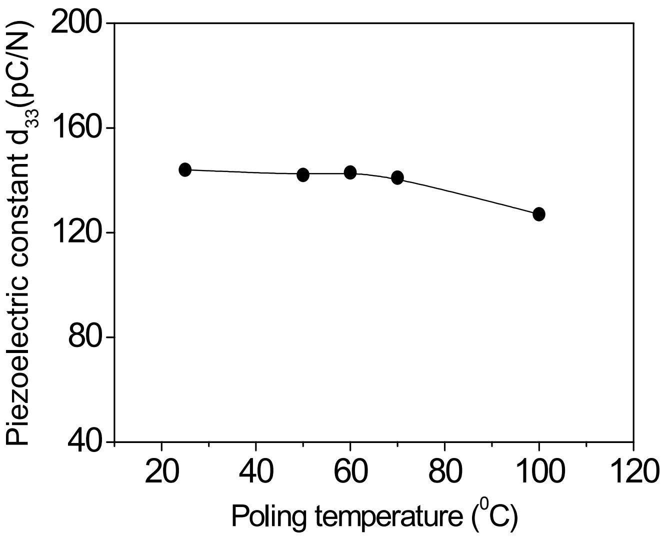

***Fig.3**: Piezoelectric constant d_{33} vs poling temperature (Poling field=50KV/cm and poling time=10min)*

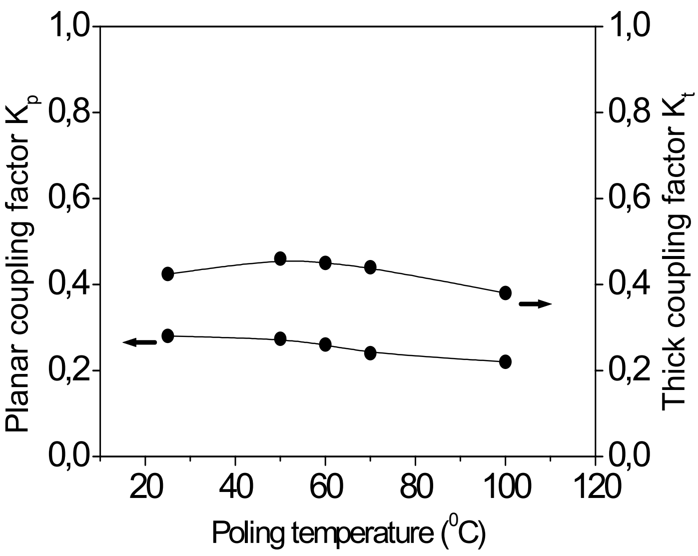

***Fig.4**: Electromechanical coupling factors (K_p and K_t) vs poling temperature of BNT-6.5BT ceramics (Poling field=50KV/cm and poling time=10min)*

From fig. 3, we can show that the piezoelectric constant d_{33} is almost constant in the range of 50-70°C for the specimens and then decreases in the range of 70-100°C of poling temperature. As the poling field is not applied during the decrease of temperature, reorientation of ferroelectric domains likely occurs for the highest temperature values i.e. 70-100°C. In the range of 50-70°C of temperature poling, the electromechanical factors (k_p and k_t) seem almost unchanged but slightly decrease with the poling temperature from 70-100 °C for the samples (fig.4). In general they display the same variation like the piezoelectric constant d_{33}.

A low value of the piezoelectric constant was observed at poling temperature of 120°C (d_{33}= 45 pC/N). This low value of the piezoelectric constant can probably allotted to the relatively low depolarization temperature of the BNT-xBT solid solution near Morphotropic Phase Boundary (MPB) [11]-[12]. 120°C is a value of temperature close to a first allotropic transition of BNT-xBT materials (ferroelectric-antiferroelectric transition) which one participates to some moderate depolarisation and then explains the slight low values of measured piezoelectric constant. In addition the specimens are easily broken down when the poling temperature is over 100°C because of the increase of electrical conductivity. With respect to that the poling temperature range 50-70 °C is choosing for our BNT-6.5BT ceramics.

A variation on the poling time in the range 5-30 min do not enhanced the piezoelectric properties under 50 KV/cm at 60°C. For example the dependency of piezoelectric constant d_{33} on poling time is shown in fig. 5.

Under poling field of 50 KV/cm at poling temperature range of 50-70 °C for 10 min, the ceramics exhibit a good piezoelectric performances: The piezoelectric coefficient d_{33}, the planar coupling factor k_p and the thick coupling factor k_t are found to be over 140 pC/N, 0.24 and 0.44 respectively and the k_t/k_p ratio ranges from 1.70 to 1.83. These relatively high piezoelectric and electromechanical activities can be attributed to MPB composition [13].

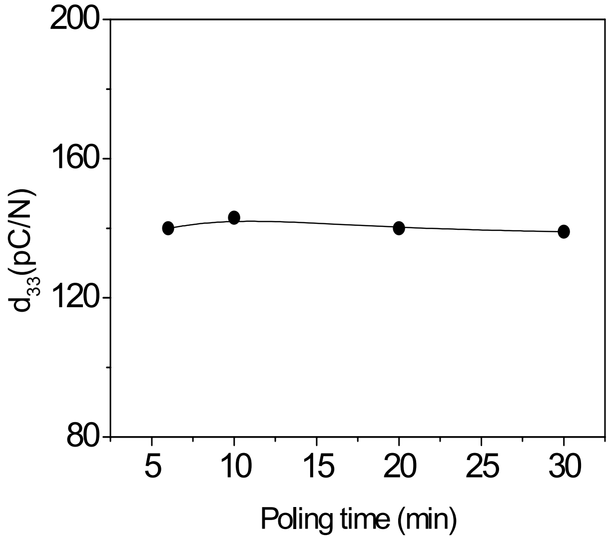

***Fig. 5**: Piezoelectric constant d_{33} vs poling time of BNT-6.5BT ceramics (Poling field=50KV/cm and poling temperature=60^{0}C).*

4. CONCLUSION

In conclusion, the effect of poling conditions on the piezoelectric properties of BNT-6.5BT ceramics was studied. It was found that the piezoelectric properties depend on poling field and poling temperature, while no remarkable effect of the poling time was detected in the range of 5-30 min. The poling temperature 50-70 ^{0}C and the poling field of 50KV/cm are the preferred poling conditions for BNT-6.5BT ceramics.

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