Effects of Uniaxial Stress on Dielectric Properties of PZT, BT and 0.55PZT-0.45BT Ceramics

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Abstract

Effects of uniaxial stress on the dielectric properties of PZT, BT and 0.55PZT-0.45BT ceramics are investigated. The ceramics are prepared by a conventional mixed-oxide method. Phase formation behavior is studied by x-ray diffraction. The dielectric properties of PZT, BT and 0.55PZT-0.45BT ceramics are observed under the uniaxial stress at low and high-stress levels using a compressometer. The results show that the dielectric constant of the ceramics at low-stress level (0-700 kPa) increases slightly, while the dielectric loss tangent changes uncertainly, with increasing applied stress. At higher stress levels (0-16 MPa), the dielectric constant and the dielectric loss tangent of PZT and 0.55PZT-0.45BT ceramics increase significantly with increasing applied stress. In the other hand, the dielectric constant and the dielectric loss tangents of BT ceramic is reduce with increasing applied stress.These results are believed to be caused by the domain structure changes to maintain the domain energy at a minimum under applied stress.

Keywords : Dielectric properties, Uniaxial stress, PZT-BT

Introduction

Lead zirconate titanate ($Pb(Zr_{1-x}Ti_x)O_3$, PZT) and barium titanate ($BaTiO_3$, BT) ceramics are widely used in devices like piezoelectric actuators and electromechanical transducers (Haertling, 1999). These two types of ceramics possess distinct characteristics that make each ceramic suitable for different applications. Though BT ceramics were the first piezoelectric transducer ceramic ever developed, it is used more widely as high-dielectric constant capacitors. BT has a low Curie temperature (T_c) of 120°C which limits its use as high-power transducers (Haertling, 1999). On the other hand, PZT ceramics have much higher Curie temperatures (T_c) (depending upon compositions), high electromechanical coupling factor (0.65), possess a wide range of dielectric constants, and form solid-solution compositions with many different constituents, thus allowing a wide range of achievable properties. However, PZT ceramics are fairly lossy as a result of their highly hysteretic behavior, which makes them unsuitable for applications that require high delicacy and reliability (Haertling, 1999). With the complementary features of PZT and BT, it is of special interest to investigate a solid solution of PZT-BT ceramics, which is expected to possess more desirable features than single-phase PZT and BT.

Furthermore, these ceramics are often subjected to external mechanical loading when used in specific applications. A prior knowledge of how the material

properties change under different load conditions is crucial for proper design of a device and for suitable selection of materials for specific applications. Despite this fact, material constants used in any design calculation are often obtained from a stress-free measuring condition, which in turn may lead to incorrect or inappropriate actuator and transducer designs. It is therefore important to determine the properties of these materials as a function of applied stress. Previous investigations on the stress-dependent dielectric and electrical properties of other ceramic systems, such as PZT and PMN-PT have clearly emphasized the importance of the subject (Zhao *et al,* 1996, Yang *et al,* 2000, Zhao *et al,* 1999). However, there has been no report on the study of PZT-BT systems. Therefore, this study is undertaken to investigate the dielectric properties of PZT, BT and 0.55PZT-0.45BT ceramics under uniaxial stress.

Methodology

In this study, BT powders were prepared from reagent grade $BaCO_3$ and TiO_2 starting powders. These powders were ball-milled for 24 h and later calcined at 1300°C for 2 h with 5°C/min heating and cooling rates. PZT powders were prepared using a lead zirconate ($PbZrO_3$) precursor in order to reduce the occurrence of undersirable pyrochlore phase (Chaisan *et al,* 2004). The perovskite phase $PbZrO_3$ was first prepared from PbO and ZrO_2 powders and then ball-milled for 24 h and calcined at 850°C for 1 h with 10°C/min heating and cooling rates. PbO and TiO_2 were then added to the calcined $PbZrO_3$ precursor, wet-mixed and calcined at 900°C for 2 h with 5°C/min heating and cooling rates. The 0.55PZT-0.45BT powder was prepared from the starting PZT and BT powders by a mixed-oxide method. The PZT and BT powders were weighed and then ball-milled for 24 h.

The PZT, BT and 0.55PZT-0.45BT ceramics were prepared from these powders pressed hydraulically under uniaxial pressure at 1.5 ton to form disc-shaped pellets 15 mm in diameter, with 3% polyvinyl alcohol (PVA) as a binder. The pellets were stacked in a covered alumina crucible filled with $PbZrO_3$ powders to prevent lead loss for PZT and 0.55PZT-0.45BT, except for BT. Finally, the sintering was carried out at a sintering temperature for 2 h with 5°C/min heating and cooling rates. The firing profile included 1 h soaking at 500°C for binder burnout process to complete. For optimization purpose, the sintering temperature was varied between 1050°C and 1250°C for PZT, between 1100°C and 1300°C for 0.55PZT-0.45BT, and between 1200°C and 1400°C for BT.

The densities of the sintered ceramics were measured by the Archimedes method. The firing shrinkage was determined from the dimensions of the specimens before and after the sintering process. The phase formations of the sintered specimens were studied by an x-ray diffractometer (JEOL Model JDX-8030). The effects of uniaxial stress on the dielectric properties were observed at low and high-stress levels with the uniaxial compressometer (Yimnirun *et al,* 2003). The dielectric properties were measured through spring-loaded pins connected to LCZ-meter (Instek MODEL LCR-821). The capacitance and the dielectric loss tangent were determined at frequency of 1 kHz and room temperature (25°C). The dielectric constant was then calculated from a parallel-plate capacitor equation, e.g.

$\varepsilon_r = Cd/\varepsilon_0 A$, where C was the capacitance of the sample, d and A were the thickness and the area of the electrode, respectively, and ε_0 was the dielectric permittivity of vacuum (8.854×10^{-12} Fm^{-1}).

Results and Discussion

The density and the firing shrinkage of sintered PZT, BT and 0.55PZT-0.45BT ceramics as a function of the sintering temperatures are shown in Fig. 1 and 2. The density of sintered PZT increases with rising sintering temperature over the range of 1050°C and 1150°C, where it reaches the maximum value. Further increasing in the sintering temperature to 1250°C results in a fall in the sintered density. This is due to lead evaporation in a form of PbO when the sintering temperature is increased (Cho *et al,* 1983). On the other hand, the densities of sintered BT and 0.55PZT-0.45BT increase with rising sintering temperature.

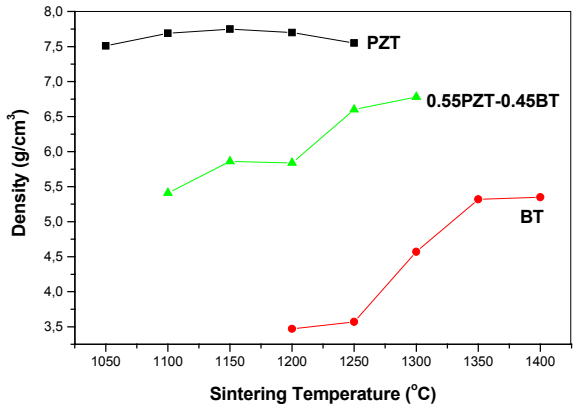

Figure 1 The density of sintered PZT, BT and 0.55PZT-0.45BT ceramics as a function of sintering temperature.

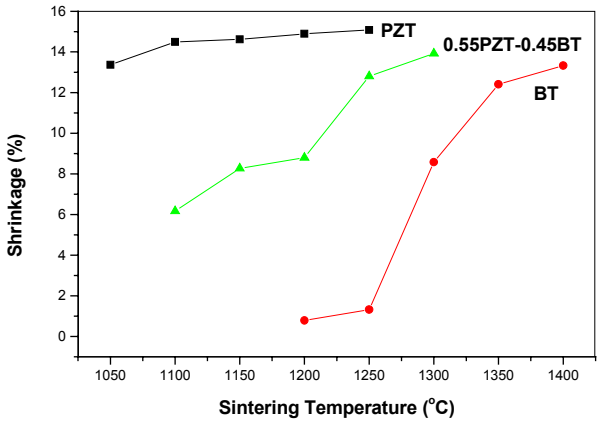

Figure 2 The shrinkage of sintered PZT, BT and 0.55PZT-0.45BT ceramics as a function of sintering temperature.

From Fig. 2, the firing shrinkage of the sintered ceramics increase with rising sintering temperature. Shrinkages of the PZT ceramic is larger than 0.55PZT-0.45BT and BT ceramics. This is due to low shrinkage of BT powder.

The phase formation behavior of the highest density sintered ceramics is revealed by an x-ray diffraction (XRD) method. The XRD patterns, shown in Fig. 3, show that the sintered ceramics are mainly in perovskite phase. From the XRD patterns, PZT and BT ceramics are identified as a single-phase material with a perovskite structure having tetragonal symmetry. The 0.55PZT-0.45BT ceramic is also in perovskite phase with tetragonal symmetry.

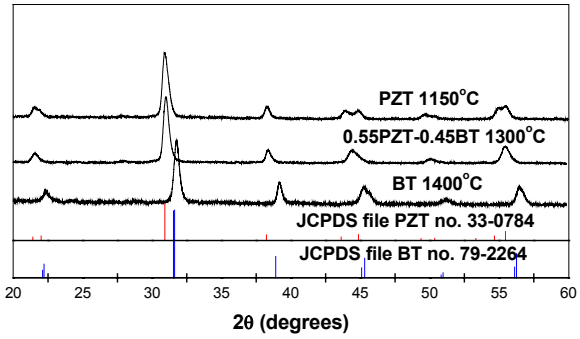

Figure 3 XRD patterns of the sintered PZT, BT and 0.55PZT-0.45BT ceramics.

The experimental results of the uniaxial stress dependence of the dielectric properties of PZT, BT and 0.55PZT-0.45BT ceramics are shown in Fig. 4 and 5 for low-stress level and Fig. 6 and 7 for high stress level. In Fig. 4 and 5, there is a slight change in the dielectric constant when the applied stress increases from 0 to 700 kPa. However, change of the dielectric loss tangent with stress is rather uncertain.

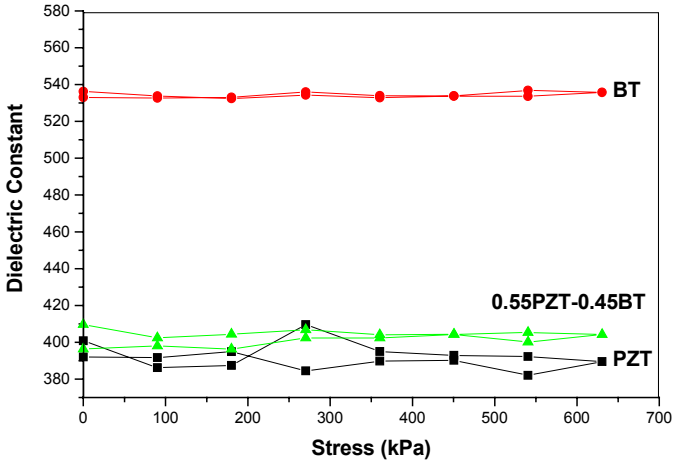

Figure 4 Uniaxial stress dependence of dielectric constant of PZT, BT and 0.55PZT-0.45BT ceramics at low-stress level.

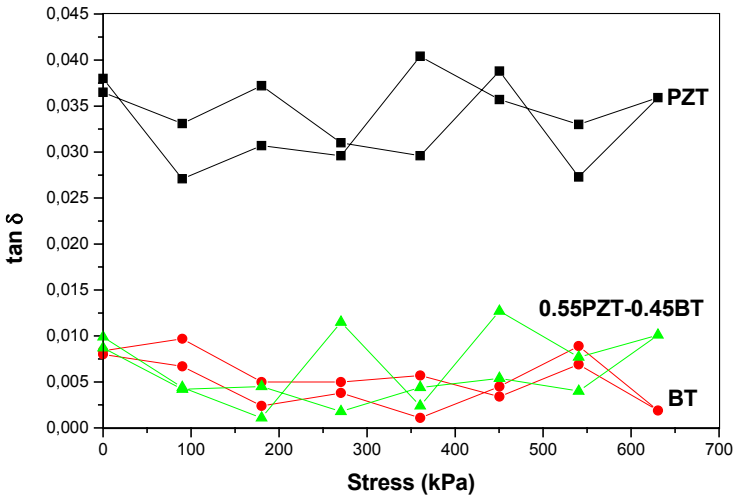

Figure 5 Uniaxial stress dependence of the dielectric loss tangent of PZT, BT and 0.55PZT-0.45BT ceramics at low-stress level.

For high-stress level, shown in Fig. 6 and 7, the changes of both the dielectric constant and the dielectric loss tangent are very significant when the applied stress increases from 0 to 16 MPa. It can be seen very clearly that the dielectric properties increase with stress for PZT and 0.55PZT-0.45BT ceramics, except for BT ceramic.

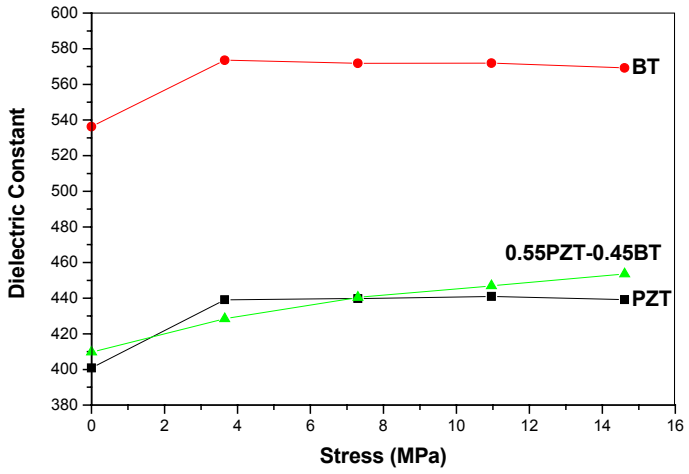

Figure 6 Uniaxial stress dependence of the dielectric constant of PZT, BT and 0.55PZT-0.45BT ceramics at high-stress level.

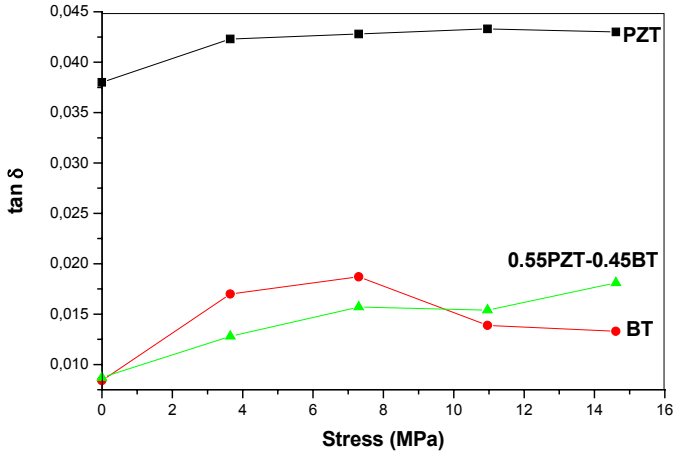

Figure 7 Uniaxial stress dependence of the dielectric loss tangent of PZT, BT and 0.55PZT-0.45BT ceramics at high-stress level.

To understand these experimental results, the properties of ferroelectric materials are derived from both the intrinsic contribution, which is response from a single domain, and extrinsic contributions, which are from domain wall motions (Yang *et al,* 2000, Zhang *et al,* 1997). When a mechanical stress is applied to a ferroelectric material, the domain structure in the material will change to maintain the domain energy at a minimum. Under a uniaxial stress, the domain structure of ferroelectric ceramics may undergo domain switching, clamping of domain walls, de-aging and de-poling (Yang *et al,* 2000).

Conclusions

The dielectric properties under the uniaxial stress of the PZT, BT and 0.55PZT-0.45BT ceramics are observed at stress levels from 0 to 700 kPa and from 0 to 16 MPa with the uniaxial compressometer. The results show that the dielectric constant at low-stress levels increase slightly with increasing applied stress while the dielectric loss tangent change are uncertain. At high-stress levels, both the dielectric constant and the dielectric loss tangents increase significantly with increasing applied stress for PZT and 0.55PZT-0.45BT. In the other hand, the dielectric constant and the dielectric loss tangents of BT ceramic is reduce with increasing applied stress. This is due to the domain structure changes to maintain the domain energy at a minimum under applied stress.

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