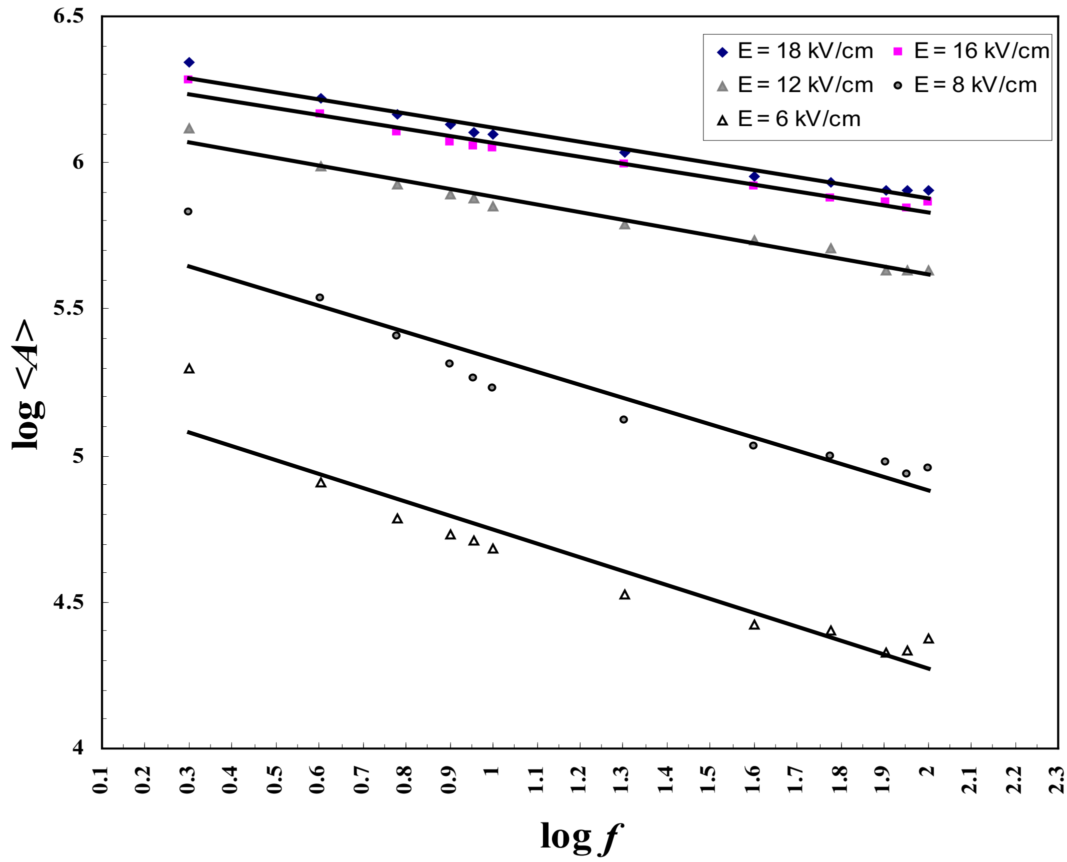
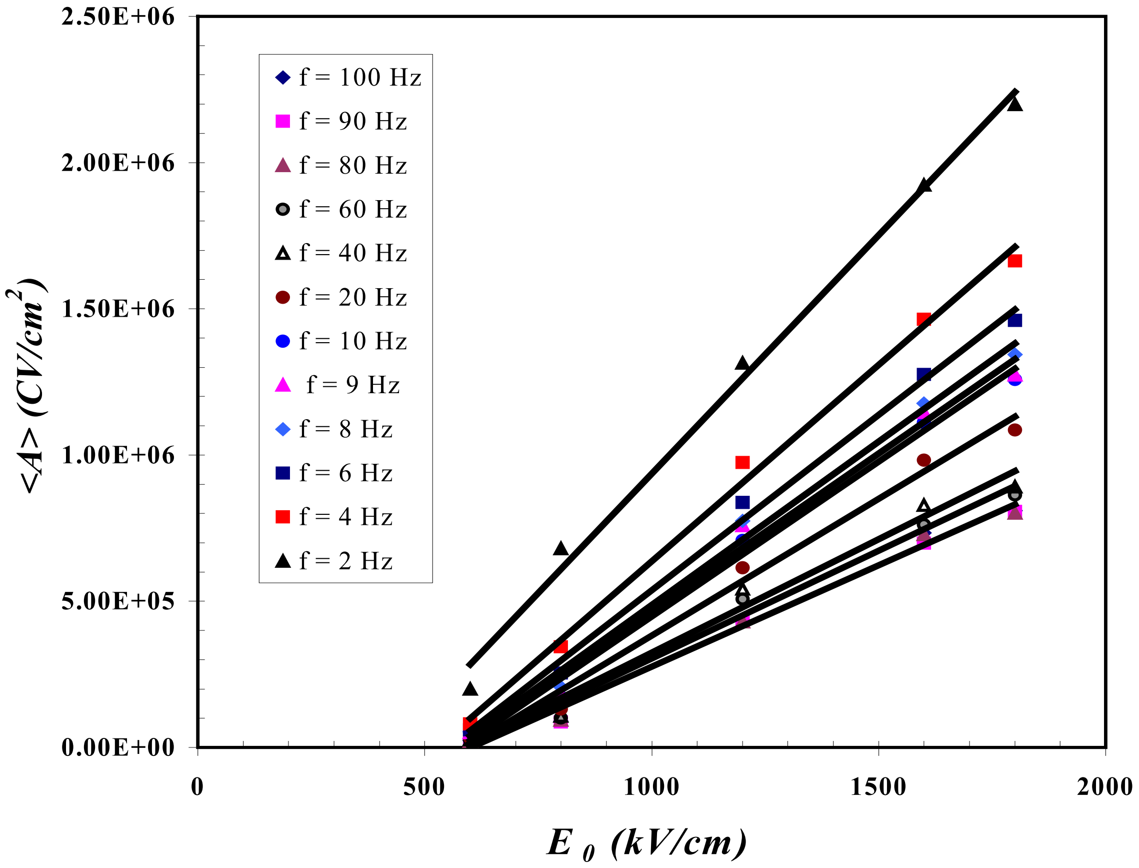
Fig. 3(a)



◆ E = 18 kV/cm
■ E = 16 kV/cm
▲ E = 12 kV/cm
○ E = 8 kV/cm
△ E = 6 kV/cm
log <*A*>
6.5
6
5.5
5
4.5
4
log *f*
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
1.1
1.2
1.3
1.4
1.5
1.6
1.7
1.8
1.9
2
2.1
2.2
2.3

Fig. 3(b)


2.50E+06
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1.50E+06
1.00E+06
5.00E+05
0.00E+00
<A> (CV/cm²)
0
500
1000
1500
2000
E 0 (kV/cm)
f = 100 Hz
f = 90 Hz
f = 80 Hz
f = 60 Hz
f = 40 Hz
f = 20 Hz
f = 10 Hz
f = 9 Hz
f = 8 Hz
f = 6 Hz
f = 4 Hz
f = 2 Hz

Fig. 3(c)

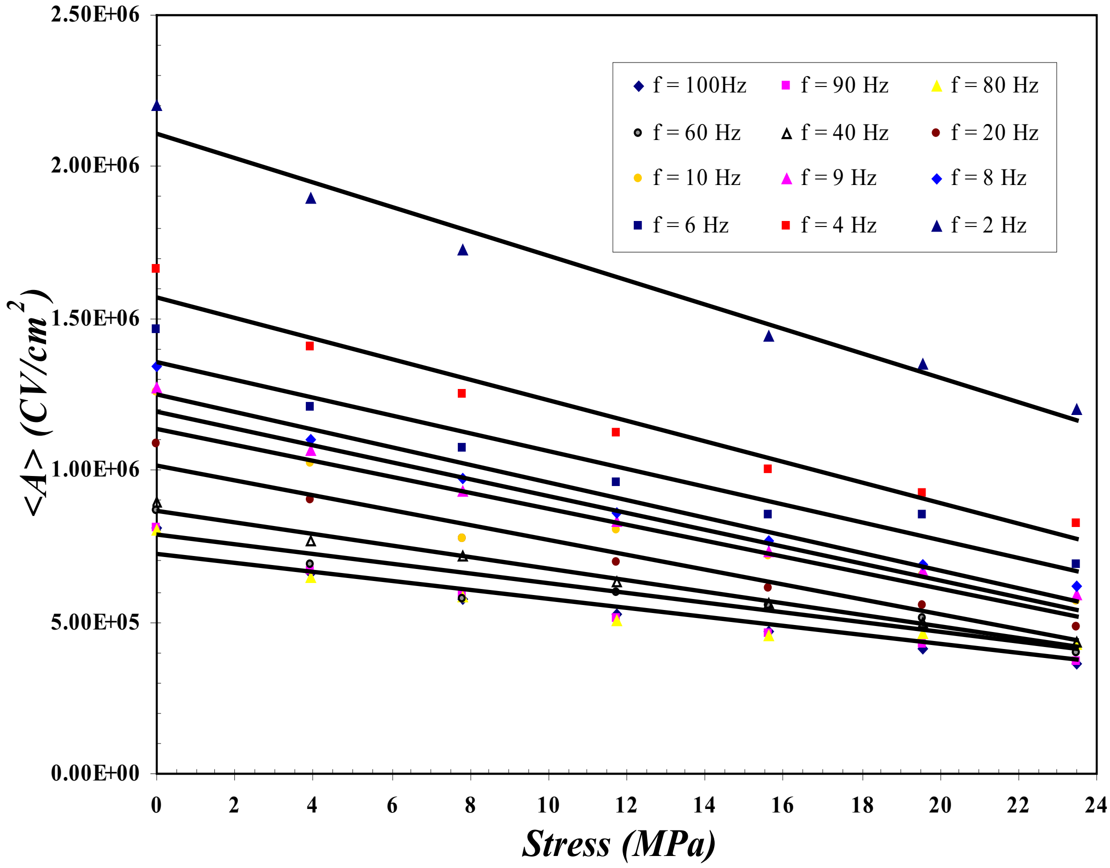
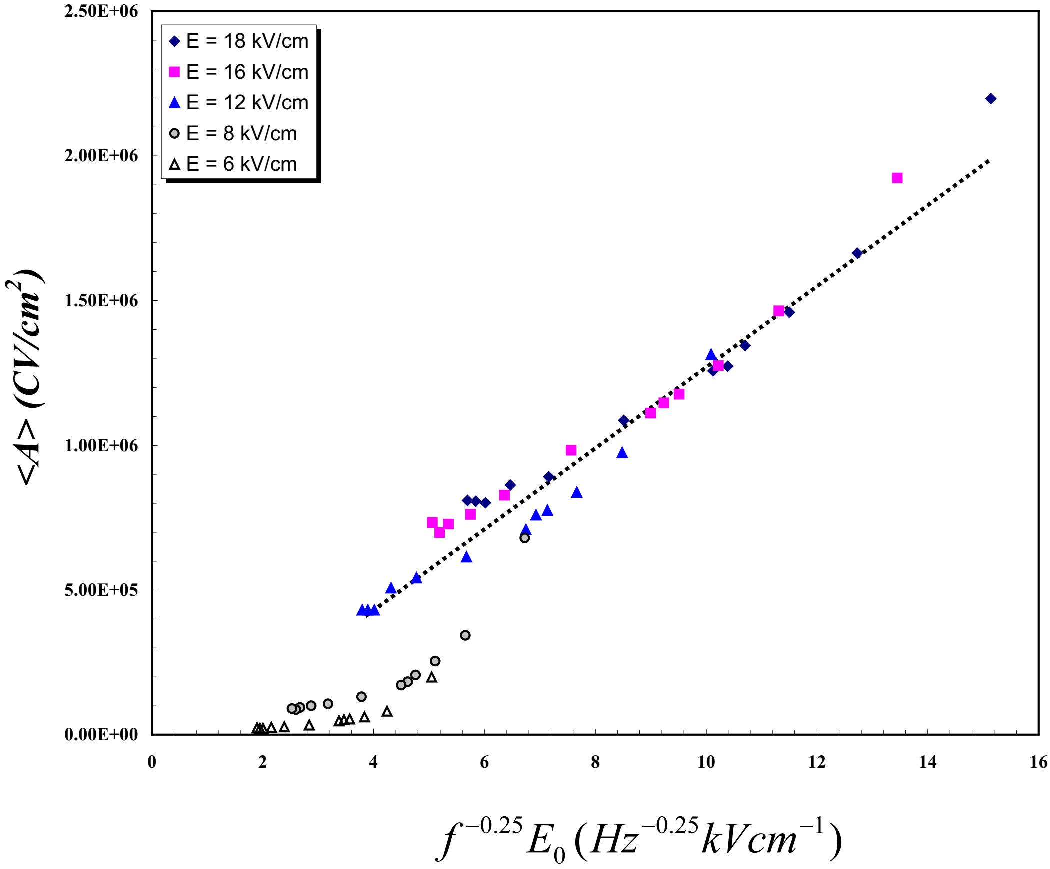

2.50E+06
2.00E+06
1.50E+06
1.00E+06
5.00E+05
0.00E+00
<A> (CV/cm^2)
0
2
4
6
8
10
12
14
16
18
20
22
24
Stress (MPa)
f = 100Hz
f = 90 Hz
f = 80 Hz
f = 60 Hz
f = 40 Hz
f = 20 Hz
f = 10 Hz
f = 9 Hz
f = 8 Hz
f = 6 Hz
f = 4 Hz
f = 2 Hz

Fig. 4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E = 16 kV/cm
E = 12 kV/cm
E = 8 kV/cm
E = 6 kV/cm
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2.00E+06
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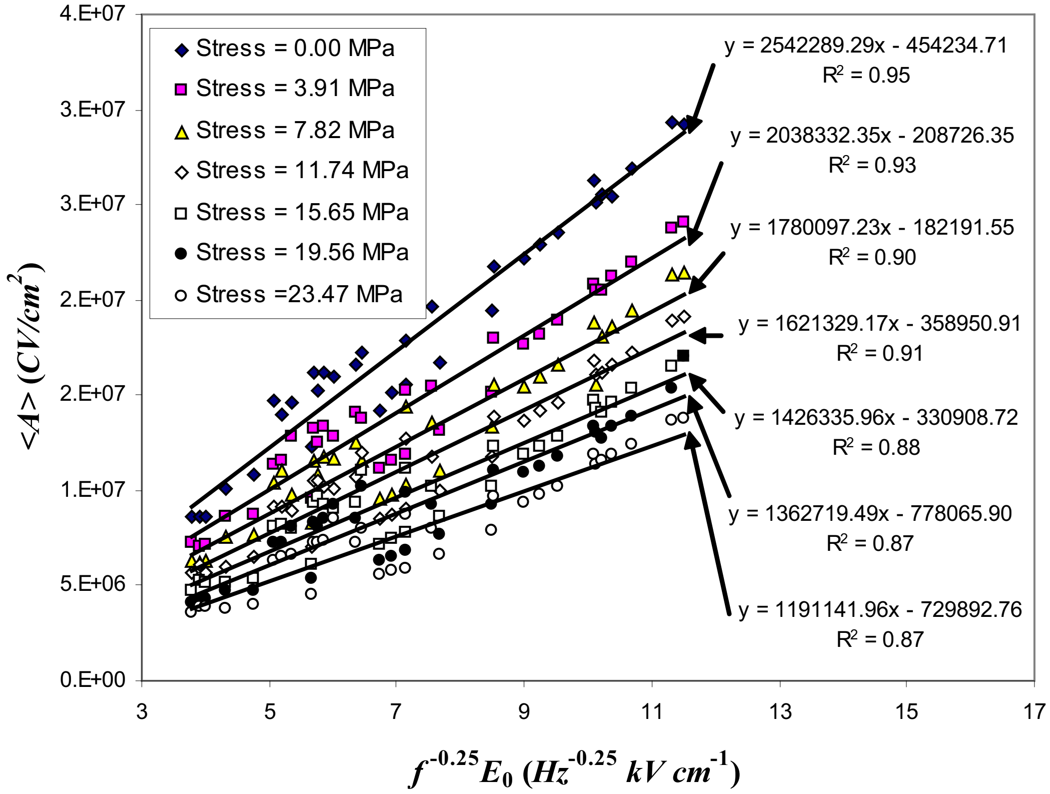
1.00E+06
5.00E+05
0.00E+00
0
2
4
6
8
10
12
14
16
$<A>\ (CV/cm^2)$
$f^{-0.25}E_0\,(Hz^{-0.25}kVcm^{-1})$

Fig. 5



4.E+07
3.E+07
3.E+07
3.E+07
2.E+07
2.E+07
1.E+07
5.E+06
0.E+00
3
5
7
9
11
13
15
17
◆ Stress = 0.00 MPa
■ Stress = 3.91 MPa
▲ Stress = 7.82 MPa
◇ Stress = 11.74 MPa
□ Stress = 15.65 MPa
● Stress = 19.56 MPa
○ Stress =23.47 MPa
y = 2542289.29x - 454234.71
R² = 0.95
y = 2038332.35x - 208726.35
R² = 0.93
y = 1780097.23x - 182191.55
R² = 0.90
y = 1621329.17x - 358950.91
R² = 0.91
y = 1426335.96x - 330908.72
R² = 0.88
y = 1362719.49x - 778065.90
R² = 0.87
y = 1191141.96x - 729892.76
R² = 0.87
<A> (CV/cm²)
f^-0.25 E0 (Hz^-0.25 kV cm^-1)

Fig. 6

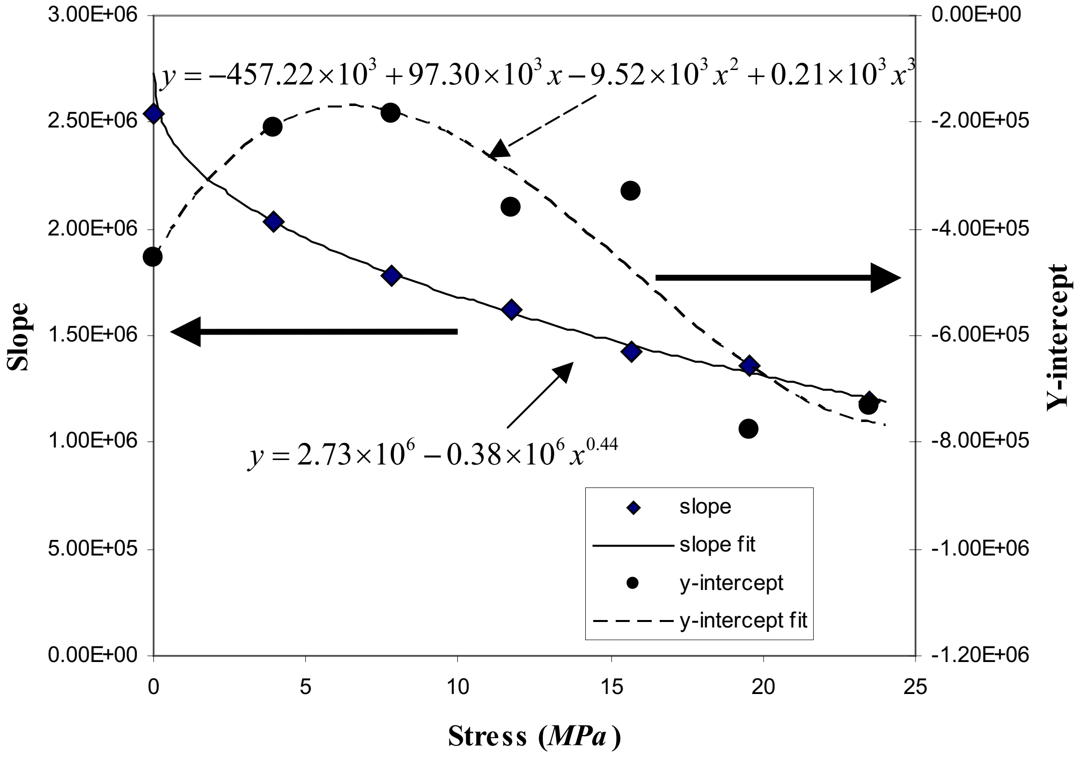
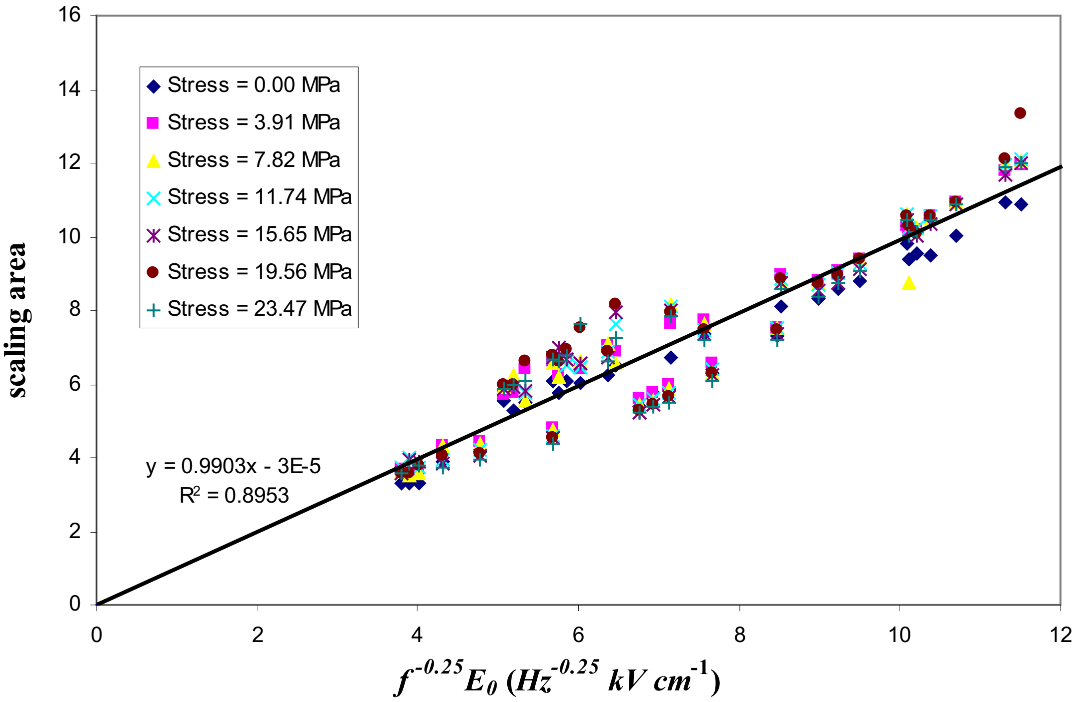

3.00E+06
2.50E+06
2.00E+06
1.50E+06
1.00E+06
5.00E+05
0.00E+00
Slope
0.00E+00
-2.00E+05
-4.00E+05
-6.00E+05
-8.00E+05
-1.00E+06
-1.20E+06
Y-intercept
0
5
10
15
20
25
Stress (MPa)
$y=-457.22\times10^{3}+97.30\times10^{3}x-9.52\times10^{3}x^{2}+0.21\times10^{3}x^{3}$
$y=2.73\times10^{6}-0.38\times10^{6}x^{0.44}$
slope
slope fit
y-intercept
y-intercept fit

Fig. 7


16
14
12
10
8
6
4
2
0
0
2
4
6
8
10
12
scaling area
f^{-0.25}E_0 (Hz^{-0.25} kV cm^{-1})
◆ Stress = 0.00 MPa
■ Stress = 3.91 MPa
▲ Stress = 7.82 MPa
× Stress = 11.74 MPa
✳ Stress = 15.65 MPa
● Stress = 19.56 MPa
+ Stress = 23.47 MPa
y = 0.9903x - 3E-5
R² = 0.8953

Effects of Uniaxial Stress on Dielectric Properties of 0.9PMN-0.1PT Ceramics

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ABSTRACT

This study deals with the influence of uniaxial stress on the dielectric properties of electrostrictive PMN-PT ceramic. The dielectric properties of lead magnesium niobate-lead titanate prepared by a mixed-oxide method with composition ratio 9:1 were measured under uniaxial compressive stress up to 22 MPa. The experimental results revealed that the superimposed compression load significantly reduced both the dielectric constant and the dielectric loss tangent in every measuring frequency. The observations were interpreted in terms of clamping of domain walls and de-poling under the compressive loading. The change of the dielectric properties with stress was attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. In addition, the results reported here also suggested a significant influence of the experimental conditions on the uniaxial stress dependence of dielectric properties of the PMN-PT ceramic.

Keywords: uniaxial stress; dielectric properties; 0.9PMN-0.1PT

INTRODUCTION

Ferroelectric lead magnesium niobate, $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN), is widely used in devices such as actuators and transducers because of its good dielectric properties (for instance, at 100 Hz ;ε_r at room temperature ~ 13000 and ε_{max} ~ 16000) [1-3]. However, the temperature related to the maximum dielectric constant (T_{max}) of PMN is very low (~ -10 C) [3]. Thus, lead titanate ($PbTiO_3$ or PT), which has high Curie temperature (T_c ~ 490 C, ε_r at room temperature ~ 300) [3-5] is added to PMN with ratio 9:1 to enhance the dielectric properties of PMN (as well as increasing T_{max}). As a result, at 100Hz, 0.9PMN-0.1PT ceramic has T_{max} = 40 C and ε_{max} > 20000 [6]. Therefore, 0.9PMN-0.1PT ceramics have been a subject of many investigations and, more importantly, have been widely applied in actuators and transducers [1, 4, 7].

However, when used in devices specified above these ceramics are often subjected to self-induced or external stress, e.g. acoustic transducers [1,3,7]. Therefore, it is very important to obtain experimental data, as well as to better understand how these materials behave under stress. Recently, the uniaxial stress dependence of dielectric properties has been studied in materials such as PZT, PMN-PZT [8-12]. For PZT ceramics, the dielectric constant increases with increasing stress in range 0-35 MPa. After reaching a maximum value of dielectric constant at 35 MPa, it gradually decreases with further pre-stress increment [8,10,11]. On the other hand, in case of PMN-PZT ceramics, the changes of the dielectric constant with the applied stress can be divided into two different groups. For PMN-rich compositions, the dielectric constant generally decreases with increasing applied stress, while for PZT-rich compositions the dielectric constant rises slightly when the applied stress increases from 0 to 1 MPa, and becomes relatively constant when the applied stress increases further [9]. These results clearly show that the effects of stress on the dielectric properties depend significantly on ceramic compositions and stress levels. Since PMN-PT ceramics are practically very important and there have been previous reports on the electro-mechanical properties of these ceramics under various mechanical and electrical loading conditions [13,14]. However, there has been no systematic study on the influence of an applied stress on their dielectric properties. Therefore, it is the aim of this study to determine the dielectric properties of the 0.9PMN-0.1PT ceramic as a function of uniaxial compressive stress.

EXPERIMENTAL METHOD

0.9PMN-0.1PT ceramics were prepared from starting PMN and PT powders by a mixed-oxide method. Perovskite-phase PMN powders were obtained from the columbite method, while PT powders were prepared by a simple mixed-oxide method.

To obtain the perovskite-phase PMN, the magnesium niobate powders were first prepared by mixing MgO (99.0%) and Nb_2O_5 (99.9%) powders and then calcining the mixed powders at 1100 C for 3 h, to yield a so called columbite powders ($MgNb_2O_6$). After that, the columbite powders were mixed with PbO (99.9%) by ball-milling method and calcined at 800 C for 1 h to form the perovskite-phase PMN powders. With a simple mixed-oxide route, PT powders were prepared from PbO (99.9%) and TiO_2 (99.9%) starting powders. These powders were ball-milled and calcined at 600 C for 1 h.

0.9PMN-0.1PT ceramics were prepared from starting PMN and PT powders by the same mixed-oxide method with ratio 9:1. After mixing the powders by ball-milling method and drying process, the mixed powders were pressed hydraulically to form disc-shaped pellets 12 mm in diameter and 1.5 mm thick, with 3 wt.% polyvinyl alcohol as a binder. The pellets were placed on the alumina powder-bed inside alumina crucible and surrounded with atmosphere powders of the same composition. Finally, the pellets were sintered at 1240 C for 2 h. The phase-formation of the sintered ceramics was studied by XRD technique. The microstructure analyses were undertaken by a scanning electron microscopy (SEM: JEOL Model JSM 840A). The densities of sintered specimens were measured by Archimedes method. The detailed descriptions of the ceramic processing and characterization were presented elsewhere and will not be discussed here [15].

Before studying the dielectric properties under the uniaxial stress, the specimens were lapped to obtain parallel faces. After coating with silver paint as electrode at the faces, the specimens were heated at 750 C for 12 min to ensure the contact between the electrode and surface of ceramic. To study effects of the uniaxial stress on the dielectric properties of the ceramic, the uniaxial compressometer was constructed [9], as shown schematically in Figure 1. The specimen was laid between uniaxial stress rams. The dielectric properties were measured by LCR-meter (Instrek LCR-821). The room temperature (27 C) capacitance and the dielectric loss tangent were obtained at frequency range 1 to 200 kHz under uniaxial compressive pre-stress levels up to 22 MPa. The dielectric constant was then calculated from a parallel-plate capacitor equation, e.g. $_r$ = Cd / $_0$A, where C is the capacitance of the specimens, d and A are, respectively, the thickness and the area of the electrode, and $_0$ is the dielectric permittivity of vacuum (8.854×10^{-12} F/m).

RESULTS AND DISCUSSION

The dielectric constant at various frequencies of 0.9PMN-0.1PT ceramics as a function of compressive stress during loading and unloading are depicted in Figure 2. There is a significant change of the dielectric constant of the ceramic with increasing stress from 0 to 22 MPa and returning to stress-free condition. The dielectric constant decreases monotonically with increasing the compressive stress, then increases only slightly when the compressive stress is gradually removed. The changing of the dielectric constant with increasing and decreasing the applied stress does not follow the same path. In every frequency, the dielectric constant with increasing the compressive stress is larger in value than that with decreasing stress. It is also of interest to see that the stress-free dielectric constant value decreases significantly after a stress cycle. Furthermore, as shown in Figure 2, the dielectric constant at a constant stress also changes with frequency. The dielectric constant decreases significantly when the measuring frequency increases from 1 to 200 kHz. The trend is opposite for the dielectric loss tangent (as shown in Figure 3). This is a typical characteristic of a relaxor ferroelectric, in which below the dielectric maximum temperature, the dielectric constant decreases and the dielectric loss tangent increases with increasing frequency [1-4].

As displayed in Figure 3, the results of the uniaxial compressive stress dependence of the dielectric loss tangent show a similar tendency to those of dielectric constant. At each measuring frequency, the dielectric loss tangent deceases with increasing the compressive stress and then slightly increases when the compressive stress is removed (shown in Figure 3). The dielectric loss tangent is also found to decrease significantly after a stress cycle. Since the dielectric properties change with the applied stress in very similar trend for every measuring frequency, the data at 10 kHz are selected as representatives for better comparison in Figure 4 for the fractional changes of the dielectric properties with the compressive stress. It can be seen very clearly that the dielectric constant decreases as much as 70% at the maximum applied stress and only returns to slightly less than 50% of its original value when the stress is removed. Though following the same trend, the change in the dielectric loss tangent value is less significant, as it only decreases about 50% at the maximum stress and almost returns on its original value after a stress cycle. It is also noticed that the results of this study are in parts similar to the experimental results for PMN-PZT system in earlier investigation [9]. For 0.9PMN-0.1PZT ceramic, the dielectric constant generally decreases with increasing applied stress. However, the trend for change of the dielectric loss tangent is different. The dielectric loss tangent of 0.9PMN-0.1PZT is found to first increase when the applied stress is raised from 0 to 1 MPa, and then decrease with further increasing stress [9]. It is interesting to observe that a mixture of different normal and relaxor ferroelectrics responds to the applied stress in a similar manner.

To understand these experimental results, various effects have to be considered. Normally, the properties of ferroelectric materials are derives from both the intrinsic and extrinsic contribution [9-11]. When a compressive stress is applied to the ferroelectric materials, the domain structure in the material will change to maintain the domain energy at a minimum; during this process some of the domains engulf other domains or change shape irreversibly. Under a uniaxial stress, the domain structure of ferroelectric ceramics may undergo domain switching through non-180 domain walls, de-aging, de poling and clamping of domain walls [11,14].

In this study, since 0.9PMN-0.1PT is a relaxor ferroelectric with T_m ~ 39-40 C and the experiment was carried out at room temperature (~27 C) which is slightly below the T_m, the experimental observation, which shows decreases in both dielectric constant and dielectric loss tangent with increasing stress, can be attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. Since the behavior of 0.9PMN-0.1PT depends on the ratio between the micro-polar region and the non-polar matrix, in this case the micro-polar regions dominate [9-12]. Hence, the dielectric responses of the 0.9PMN-0.1PT ceramic are observed to decrease with increasing the compressive stress, as seen in Figures 2-4.

In addition, the de-poling mechanism also plays a role in the experimental results. The decrease of both the dielectric constant and the dielectric loss tangent after a stress cycle is believed to be the direct contribution of the partial de-poling effect under the compressive stress cycle [9-11].

More interestingly, earlier work by Viehland *et al.* [13] reported the large-signal polarization-electric field (P-E) relation under uniaxial stress of 0.9PMN-0.1PT ceramic. For comparison, one would be able to estimate from the P-E loops presented in that study the dielectric permittivity by using Eq. (1), which gives

$$\frac{P}{E} \qquad (1)$$

where P is the polarization difference between +2.5 and 0 kV/cm. The calculated dielectric permittivity can be called *differential permittivity*, which includes the reversible (intrinsic dielectric property) and irreversible (extrinsic domain switching related property) contributions of the materials [8,16]. The calculation shows that the differential permittivity decreases approximately 13% when the applied stress is increased from 0 to 30 MPa. This change is significantly lower than that observed in our low-field study (a decrease of more than 70% over the same stress range). Even though the reasons for the difference are still not clearly known, this observation clearly signifies the importance of the experimental conditions used to determine the dielectric properties, as well as the stress-dependence dielectric properties. More detailed study is underway and further results will be presented in future publications. Finally, the information obtained in this current work is practically very useful for design and calculation consideration of devices in low-field applications.

CONCLUSIONS

In this study, the dielectric properties of 0.9PMN-0.1PT ceramics prepared by a conventional mixed-oxide method are measured under the compressive stress from 0 to 22 MPa. The results clearly show that the dielectric constants and the dielectric loss tangent of the 0.9PMN-0.1PT ceramic decrease with increasing the compressive stress. The change of the dielectric properties with stress is attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. In addition, the difference in the uniaxial stress dependence of the dielectric properties measured under low- and high-field conditions is also apparent. Finally, this study undoubtedly shows that the applied stress has significant influences on the dielectric properties of the 0.9PMN-0.1PT ceramics.

ACKNOWLEDGEMENTS

This work is supported by the Thailand Research Fund (TRF). Additional supports from Faculty of Science and Graduate School of Chiang Mai University are gratefully acknowledged. One of the authors (N.T.) also wishes to thank the Development and Promotion of Science and Technology Talents Project of Thailand (DPST) for financial support during his study.

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List of Figure Captions

Figure 1. Schematic diagram of experimental set up.

Figure 2. Uniaxial compressive stress dependence of dielectric constant ($_r$) of 0.9PMN-0.1PT ceramic at various frequencies.

Figure 3. Uniaxial compressive stress dependence of dielectric loss tangent (tan) of 0.9PMN-0.1PT ceramic at various frequencies.

Figure 4. Fractional changes of dielectric constant ($_r$) and dielectric loss tangent (tan) with uniaxial compressive stress of 0.9PMN-0.1PT ceramic (measured at 10 kHz).

Figure 1

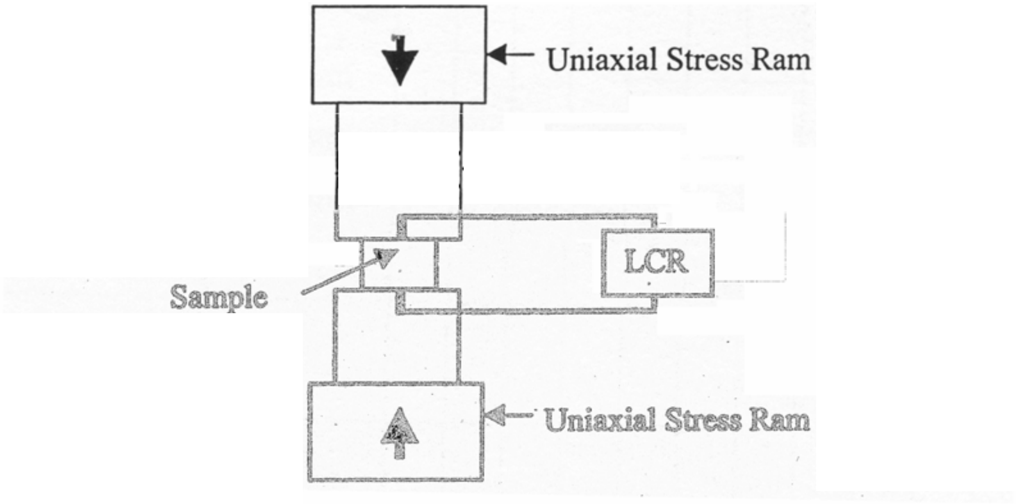

Figure 1. Schematic diagram of experimental set up.

Figure 2

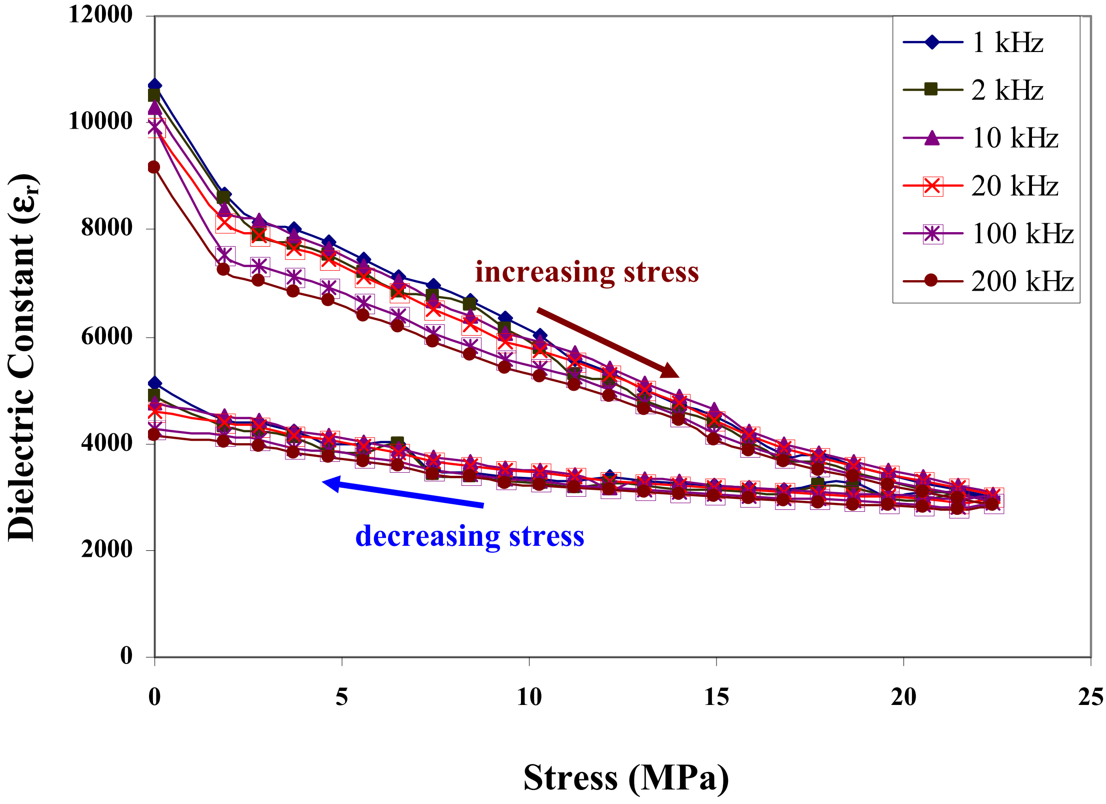

Figure 2. Uniaxial compressive stress dependence of dielectric constant ($_r$) of 0.9PMN-0.1PT ceramic at various frequencies.

Figure 3

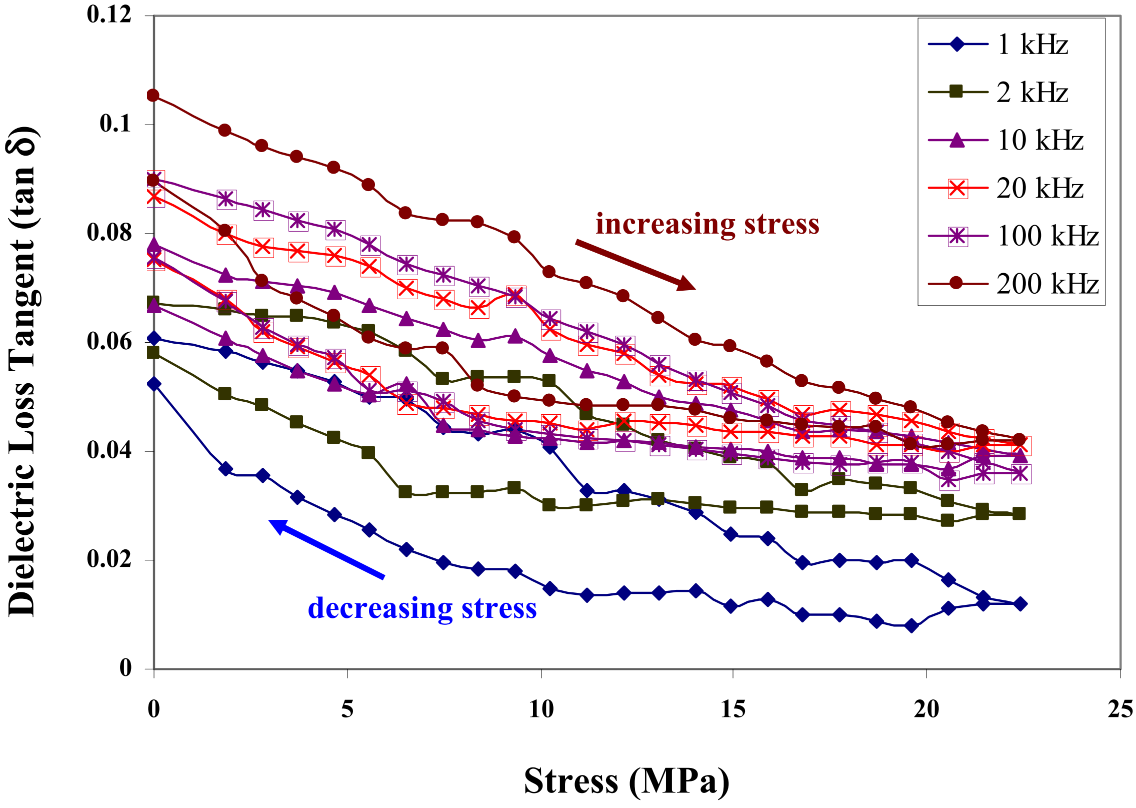

Figure 3. Uniaxial compressive stress dependence of dielectric loss tangent (tan)

of 0.9PMN-0.1PT ceramic at various frequencies.

Figure 4

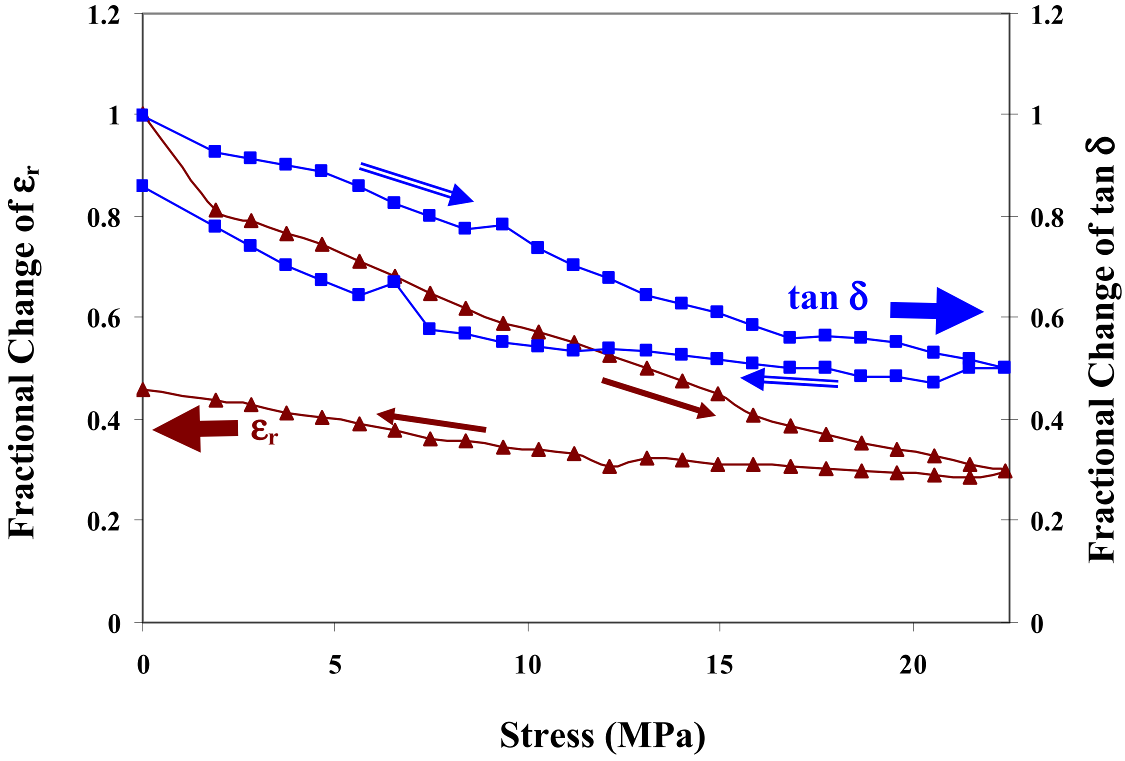

Figure 4. Fractional changes of dielectric constant ($_r$) and dielectric loss tangent (tan) with uniaxial compressive stress of 0.9PMN-0.1PT ceramic (measured at 10 kHz).

Dielectric Properties of $(1\text{-}x)\mathrm{Pb(Zr_{0.52}Ti_{0.48})O_3}$-$(x)\mathrm{BaTiO_3}$ Ceramics Under Uniaxial Compressive Pre-Stress

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ABSTRACT

Effects of uniaxial compressive pre-stress on the dielectric properties of ceramics in PZT-BT solid solution system are investigated. The physical properties measurements reveal that the addition of BT into PZT results in the increase of grain size and microstructural heterogeneity, except for the density which sees the opposite trend. The dielectric properties measured under stress-free condition show a gradual increase of the dielectric constant with increasing BT content, while the dielectric loss tangent is not considerably different. The dielectric properties under the uniaxial compressive pre-stress of the PZT-BT ceramics are observed at stress levels up to 15 MPa using a uniaxial compressometer. The results clearly show that both the dielectric constant and the dielectric loss tangent of the PZT-BT ceramics increase significantly with increasing applied stress. Larger changes in the dielectric properties with the applied stress are observed in the PZT-rich compositions. The experimental observations have been attributed to the increase of the domain wall motions from the application of the compressive pre-stress.

Keywords: Uniaxial Stress, Dielectric Properties, PZT-BT

1. Introduction

Along with nanotechnology, smart materials and systems have recently been identified as the next generation technology [1-3]. Among many smart systems, piezoelectric ceramic actuators and transducers are finding an increasingly wide range of applications. In most applications, ceramics are often subjected to mechanical loading, either deliberately in the design of the device itself or because the device is used to change shapes as in many smart structure applications or the device is used under environmental stresses. In some cases, due to the piezoelectric nature of a material used within, the devices are also subjected to the self-induced internal stress [2-4]. A prior knowledge of how the materials behave under different load conditions is therefore very crucial for proper design of a device and for suitable selection of materials for a specific application. Therefore, it is very important to determine the properties of these materials as a function of applied stress. Previous investigations on the stress-dependence dielectric and electrical properties of many ceramic systems, such as PZT, PMN-PZT, and PMN-PT have clearly emphasized the importance of the subject [4-11].

Among perovskite ferroelectric materials, lead zirconate titanate ($Pb(Zr_{1-x}Ti_x)O_3$ or PZT) and barium titanate ($BaTiO_3$ or BT) ceramics have been investigated extensively and continuously since the late 1940s [12-15]. PZT and BT are representative perovskite piezoelectric and ferroelectric prototypes, respectively, because of their excellent electrical properties. These two ceramics possess distinct characteristics that make each ceramic suitable for different applications. One of the most studied piezoelectric compounds, $Pb(Zr_{0.52}Ti_{0.48})O_3$, a morphotropic phase boundary (MPB) compound of PZT, has great piezoelectric properties with a high

Curie temperature (T_C) of ~390 ºC. BT exhibits high dielectric constant and superior electrostrictive responses with a lower T_C (~ 120 ºC) [12-15]. In addition, BT is mechanically superior than PZT [16], whereas PZT ceramics can be easily sintered at temperature much lower than BT ceramics, which usually require as high sintering temperature as 1400 ºC [16]. With their complementary features, the solid solutions between PZT and BT are expected to exhibit better properties than those of the single-phase PZT and BT [12-15]. Furthermore, the properties can also be tailored over a wider range by changing the compositions to meet the strict requirements for specific applications [3, 12-15]. Prior investigations have already revealed interesting results on the structure, and the dielectric and ferroelectric properties of the PZT-BT solid solutions [16-18]. However, there has been no report on the influences of the applied stress on the electrical properties of the PZT-BT systems. Therefore, this study is undertaken to investigate the influences of the uniaxial compressive pre-stress on the dielectric properties of $(1-x)$PZT- xBT ceramic systems.

2. Experimental procedure

The (1-x)$Pb(Zr_{0.52}Ti_{0.48})O_3$ – (x)$BaTiO_3$ ceramic systems used in this study are prepared from the PZT and BT starting powders by a mixed-oxide method. The precursor PZT and BT powders are first prepared from reagent-grade PbO (99%), ZrO_2 (99%) and TiO_2 (99%), and $BaCO_3$ (99%) and TiO_2 (99%) starting powders, respectively. These powders are ball-milled for 24 hours and later calcined for 2 hours at 900 °C for PZT and at 1300 °C for BT. Subsequently, the (1-x)$Pb(Zr_{0.52}Ti_{0.48})O_3$ – (x)$BaTiO_3$ (when x = 0.0, 0.05, 0.15, 0.25, 0.35, 0.45, 0.55, 0.65, 0.75, 0.85, 0.95, and 1.0) ceramics are prepared from the starting PZT and BT powders by a mixed-oxide method at various processing conditions. Initially, the PZT and BT powders for a given composition are weighed and then ball-milled in ethanol for 24 hours. After drying process, the mixed powders are pressed hydraulically to form disc-shaped pellets 10 mm in diameter and 2 mm thick, with 5 wt.% polyvinyl alcohol (PVA) as a binder. The pellets are stacked in a covered alumina crucible filled with PZ powders to prevent lead loss. Finally, the sintering is carried out at a sintering temperature for 2 hours with 5 min/°C heating and cooling rates. The firing profile includes a 1-hour dwell time at 500 °C for binder burnout process to complete. For optimization purpose, the sintering temperature is varied between 1050 °C and 1400 °C depending upon the compositions. The densities of the sintered ceramics are measured by Archimedes method. The phase formations of the sintered specimens are studied by an x-ray diffractometer (Philips Analytical). The microstructure analyses are undertaken by a scanning electron microscopy (SEM: JEOL Model JSM 840A). Grain size is determined from SEM micrographs by a linear intercept method. The detailed descriptions of powders and ceramics processing and characterizations are presented thoroughly in the earlier publication [16].

For dielectric property characterizations under a uniaxial compressive pre-stress, the sintered samples are lapped to obtain parallel faces disc-shaped specimens with diameter of 10 mm and thickness of 2 mm, and the faces are then coated with silver paint as electrodes. The samples are then heat-treated at 750 °C for 12 min to ensure the contact between the electrodes and the ceramic surfaces. The samples are subsequently poled in a silicone oil bath at a temperature of 120 °C by applying a dc field of 20 kV/cm for 30 min. To study the effects of the uniaxial compressive pre-stress on the dielectric properties, the uniaxial compressometer is constructed [10, 11]. As shown in Fig. 1, the compressometer is developed for simultaneous applications of the mechanical stress and the electric field. The compressometer cell consisting of a cylindrical brass cell with a heavy brass base, a brass ram and a precisely guided loading platform provides true uniaxial stress during mechanical loading. The prepared specimen is carefully placed between the two alumina blocks and the electric field is applied to the specimen via the copper shims attached to the alumina blocks. With this setting, the uniaxial compressive stress is applied parallel to the electric field direction. The uniaxial compressive stress is supplied by the servohydraulic load frame and the applied stress level is monitored with the pressure gage of the load frame. Measurements are performed as a function of mechanical pre-stress applied discretely between 0 and 15 MPa. The measurements are carried out on the samples for loading and unloading conditions. Each measurement is repeated two to three times to ensure the reliability of the experimental results. During the measurements, a desired pre-stress is first applied to the sample and the dielectric properties are then recorded. The dielectric properties are measured through the LCZ-meter (Hewlett Packard, model 4276A). The capacitance and the dielectric loss tangent are determined at frequency of 1 kHz and room temperature (25 °C). The dielectric constant is then calculated from a parallel-plate capacitor equation, e.g. $\varepsilon_r = Cd / \varepsilon_0 A$, where C is the capacitance of the sample, d and A are the thickness and the area of the electrode, respectively, and ε_0 is the dielectric permittivity of vacuum (8.854×10^{-12} F/m).

3. Results and discussion

The phase formation behavior of the sintered ceramics is revealed by an XRD method. The XRD patterns, shown in Fig. 2, show that the sintered ceramics are clearly in perovskite phase. From the XRD patterns, PZT and BT ceramics are both identified as a single-phase material with a perovskite structure having tetragonal symmetry [16]. The patterns can be matched with JCPDF files no 33-0784 and 79-2264 for PZT and BT, respectively. For other PZT-BT compositions, the diffraction peaks move gradually towards higher angles with increasing BT contents, indication of smaller cell parameters. It should also be noticed that compositions with $0.45 \leq x \leq 0.75$ exhibit evidence of a possible rhombohedral structure with no clear splitting of peaks around 2θ of 43-46º. However, it is still clearly seen from the XRD patterns that PZT-BT forms a series of continuous solid solutions [16].

The optimized density of sintered (1-x)PZT–xBT ceramics is listed in Table 1. It is observed that an addition of BT to the PZT-BT compositions results in a continual decrease in the density of the ceramics from, in the units of g/cm^3, 7.75 for PZT to 5.35 for BT. This is clearly due to the fact that BT has lower density than PZT. Except for BT which has a density of 89% of the theoretical density, all other ceramics are well sintered with high density (as high as 96% of the theoretical density in PZT ceramic). Relatively low density observed in BT ceramic is a result of porous and non-uniform microstructure, as seen from SEM micrograph in Fig. 3 (f). For BT-rich compositions, SEM micrographs also show porous microstructure which could be attributed to PbO loss during high sintering temperatures required for these compositions. In addition, an abnormal grain growth observed in the BT-rich compositions is probably due to the fact that the sintering temperatures required for highest densification for PZT and BT ceramics are very different, which could lead to distinctive grain growth behaviors between two phases [16]. Hence, the heterogeneous microstructure with a large grain size range is observed for BT-rich ceramic compositions, as seen in Figs. 3 (d-e) and as listed in Table 1. On the other hand, PZT-rich compositions exhibit more uniform microstructure with the average grain size in the range of 1-3 μm. Moreover, the average grain size of the whole compositional range increases significantly from < 2 μm in PZT to >10 μm in BT, as tabulated in Table 1.

The room temperature dielectric properties measured under stress-free condition are also listed in Table 1. It is clearly seen that dielectric constant (ε_r) of (1-x)PZT–xBT ceramics increases with increasing BT content. The dielectric constant increases from 813 in PZT to 1429 in BT. The increase of the dielectric constant within the compositional range is due to the high dielectric constant of BT [12-15]. When compared to PZT, higher dielectric constant observed in BT at room temperature is a result of its two ferroelectric transition temperatures at 0 and 120 °C [13-15]. However, the stress-free dielectric loss tangent (tan δ) does not change significantly with compositions. The relatively higher value of dielectric loss tangent of 0.011 for PZT is probably due to more domain wall motions expected in piezoelectric PZT as compared to piezoelectrically less active BT [6, 19, 20]. Similar dielectric behaviors have been reported in other solid-solution systems such as PZN-BT [21], PMN-PZT [22], and PNN-PZT [23].

The experimental results of the uniaxial compressive pre-stress dependence of the dielectric properties of the ceramics in PZT-BT system are shown in Figs. 4 and 5. There is a significant change of both the dielectric constant and the dielectric loss tangent of the ceramics when the applied stress increases from 0 to 15 MPa. The changes of the dielectric constant with the applied stress can be divided into three different groups. For PZT ceramic, the dielectric constant increases exponentially with applied stress. It can be seen that dielectric constant is enhanced by approximately 8%

at 15 MPa applied stress. For PZT-rich compositions (0.95PZT-0.05BT, 0.85PZT-0.15BT, 0.75PZT-0.25BT, 0.65PZT-0.35BT and 0.55PZT-0.45BT), the dielectric constant increases rather linearly with increasing applied stress. The changes in the dielectric constant between 2 to 4% at 15 MPa applied stress are obviously smaller than that observed in PZT. For BT-rich compositions (BT, 0.05PZT-0.95BT, 0.15PZT-0.85BT, 0.25PZT-0.75BT and 0.35PZT-0.65BT), the dielectric constant only rises slightly (< 2%) and in a linear manner when the applied stress increases to the maximum amplitude. The dielectric loss tangent for all compositions is found to increase significantly and non-linearly when the applied stress is raised from 0 to 15 MPa. The largest changes occur in PZT and 0.75PZT-0.25BT with the dielectric loss tangent enhancement of nearly 80% and 50%, respectively. For other compositions, the increase in the dielectric loss tangent varies between 10 and 40% at 15 MPa applied stress. Again the changes of the dielectric loss tangent of BT-rich compositions are comparatively smaller than those of PZT-rich compositions, similar to what have been observed in the case of the dielectric constant. Similar experimental results have been reported previously for soft PZT [9, 24], un-doped PZT with various Zr/Ti ratio [25], and Ca-doped BT [26], in which the dielectric properties are found to increase with increasing magnitude of the compressive pre-stress.

To understand these experimental results, various effects have to be considered. Normally, the properties of ferroelectric materials are derived from both the intrinsic contribution, which is the response from a single domain, and extrinsic contributions, which are from domain wall motions [19, 25-27]. 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; during this process some of the domains engulf other domains or change shape irreversibly. Under a uniaxial stress, the domain structure of ferroelectric ceramics may undergo domain switching, clamping of domain walls, de-aging, and de-poling [20, 27]. In this study, the results on the uniaxial compressive pre-stress dependence of the dielectric properties can easily be explained with the above statements. When the compressive uniaxial stress is applied in the direction parallel to the polar axis (poling) direction, the stress will move some of the polarization away from the poling direction resulting in a change in domain structures [6, 25-27]. This change increases the non-180º domain wall density. Hence the increase of the dielectric constant with the applied stress is observed. The de-aging mechanism is also expected to play a role here. However, the stress clamping of domain walls and the de-poling mechanisms are not expected at this relatively low stress level used in this study [6, 27]. Therefore, a combination of the domain switching and the de-aging mechanisms is believed to be a reason for the increase of the dielectric constant with increasing applied stress in the PZT-BT system, as shown in Fig. 4. The cause of the stress dependence of the dielectric loss tangent is a little more straightforward than that of the dielectric constant. Stress-induced depinning of the domain walls is expected to occur under the applied compressive pre-stress. As depicted in Fig. 5, an increase in domain wall mobility clearly enhances the dielectric loss tangent in all compositions [19, 20, 27]. Finally, it should be noted here that the dielectric behaviors under the applied stress for PZT-BT system are significantly different from those observed in a solid-solution between normal and relaxor ferroelectrics, i.e. PZT-PMN systems, in which the dielectric responses to the applied stress depend more on the compositions and the stress level, and the dielectric properties of some compositions decrease with increasing applied stress [10].

4. Conclusions

In this study, effects of uniaxial compressive pre-stress on the dielectric properties of ceramics in PZT-BT system are investigated. The (1-x)Pb($Zr_{0.52}Ti_{0.48}$)O_3 – (x)$BaTiO_3$ (when x = 0.0, 0.05, 0.15, 0.25, 0.35, 0.45, 0.55, 0.65, 0.75, 0.85, 0.95, and 1.0) ceramics used are prepared by a conventional mixed-oxide method from starting PZT and BT powders at various processing conditions. The phase formation behavior studied using the XRD indicates that PZT-BT forms a series of continuous solid solutions. The physical properties measurements reveal that the properties are relatively composition-dependent. Except for the density which sees the opposite trend, the addition of BT into PZT results in the increase of grain size and microstructural heterogeneity. The dielectric properties measured under stress-free conditions show a gradual increase of the dielectric constant with increasing BT content, while the dielectric loss tangent is not significantly different. The dielectric properties under the uniaxial compressive pre-stress of the PZT-BT ceramics are observed at stress levels up to 15 MPa using a uniaxial compressometer. The results clearly show that both the dielectric constant and the dielectric loss tangent of the PZT-BT ceramics increase significantly with increasing applied stress. Larger changes in the dielectric properties with the applied stress are observed in the PZT-rich compositions. The experimental observations have been attributed to the increase of the domain wall motions from the application of the compressive pre-stress.

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Table 1. Microstructural and dielectric features of (*1*-*x*)PZT-*x*BT ceramics

Ceramic	**Density (g/cm^3)**	**Grain Size Range (μm)**	**Average Grain Size (μm)**	**Stress-Free ε_r (25°C/1 kHz)**	**Stress-Free tan δ (25°C/1 kHz)**
PZT	7.75	1-5	1.89±0.52	813	0.011
0.95PZT-0.05BT	7.67	1-4	1.66±0.32	809	0.007
0.85PZT-0.15BT	7.42	1-5	2.40±0.58	839	0.006
0.75PZT-0.25BT	7.30	1-3	1.91±0.47	857	0.004
0.65PZT-0.35BT	6.99	1-4	2.36±0.71	875	0.005
0.55PZT-0.45BT	6.78	1-6	2.93±0.53	890	0.006
0.45PZT-0.55BT	6.63	1-7	3.17±0.72	952	0.005
0.35PZT-0.65BT	6.38	1-10	4.42±0.79	1060	0.007
0.25PZT-0.75BT	6.28	1-10	3.28±0.53	1077	0.005
0.15PZT-0.85BT	6.07	1.5-9	3.52±0.77	1223	0.004
0.05PZT-0.95BT	5.95	2-10	4.86±0.88	1288	0.005
BT	5.35	7-12	10.82±0.53	1429	0.004

List of Figure Captions

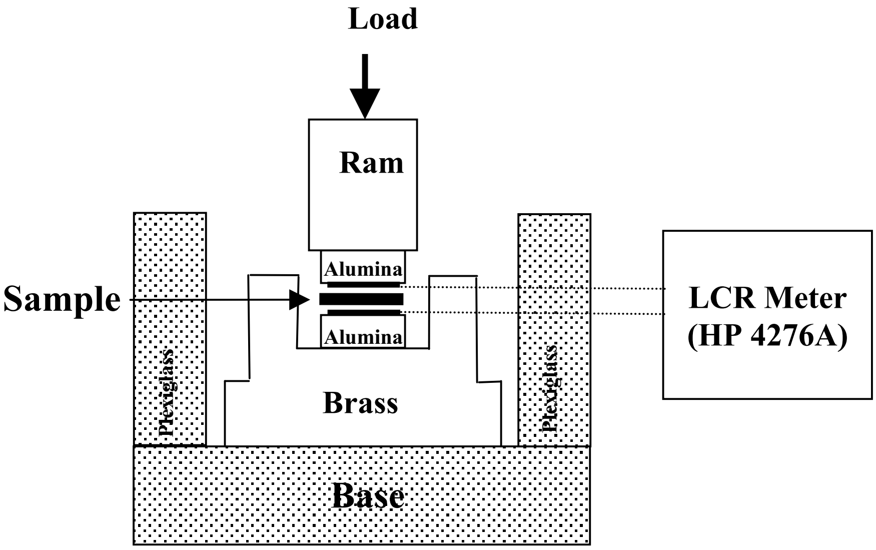
Fig. 1. A schematic of the experimental set-up

Fig. 2. XRD diffraction patterns of the sintered (*1-x*)PZT-(*x*)BT ceramics.

Fig. 3. SEM micrographs of (*1-x*)PZT-*x*BT ceramics: (a) PZT; (b) 0.75PZT-0.25BT; (c) 0.55PZT-0.45BT; (d) 0.35PZT-0.65BT; (e) 0.15PZT-0.85BT; and (f) BT.

Fig. 4. Relative changes of dielectric constant (ε_r) as a function of compressive pre-stress for (*1-x*)PZT-(*x*)BT ceramics.

Fig. 5. Relative changes of dielectric loss tangent (tan δ) as a function of compressive pre-stress for (*1-x*)PZT-(*x*)BT ceramics.



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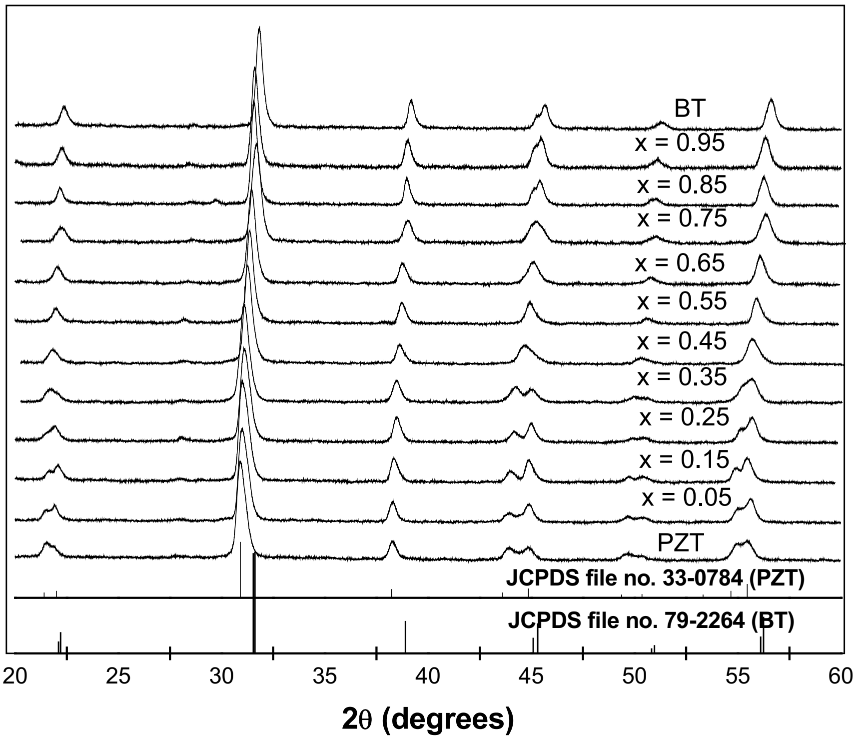

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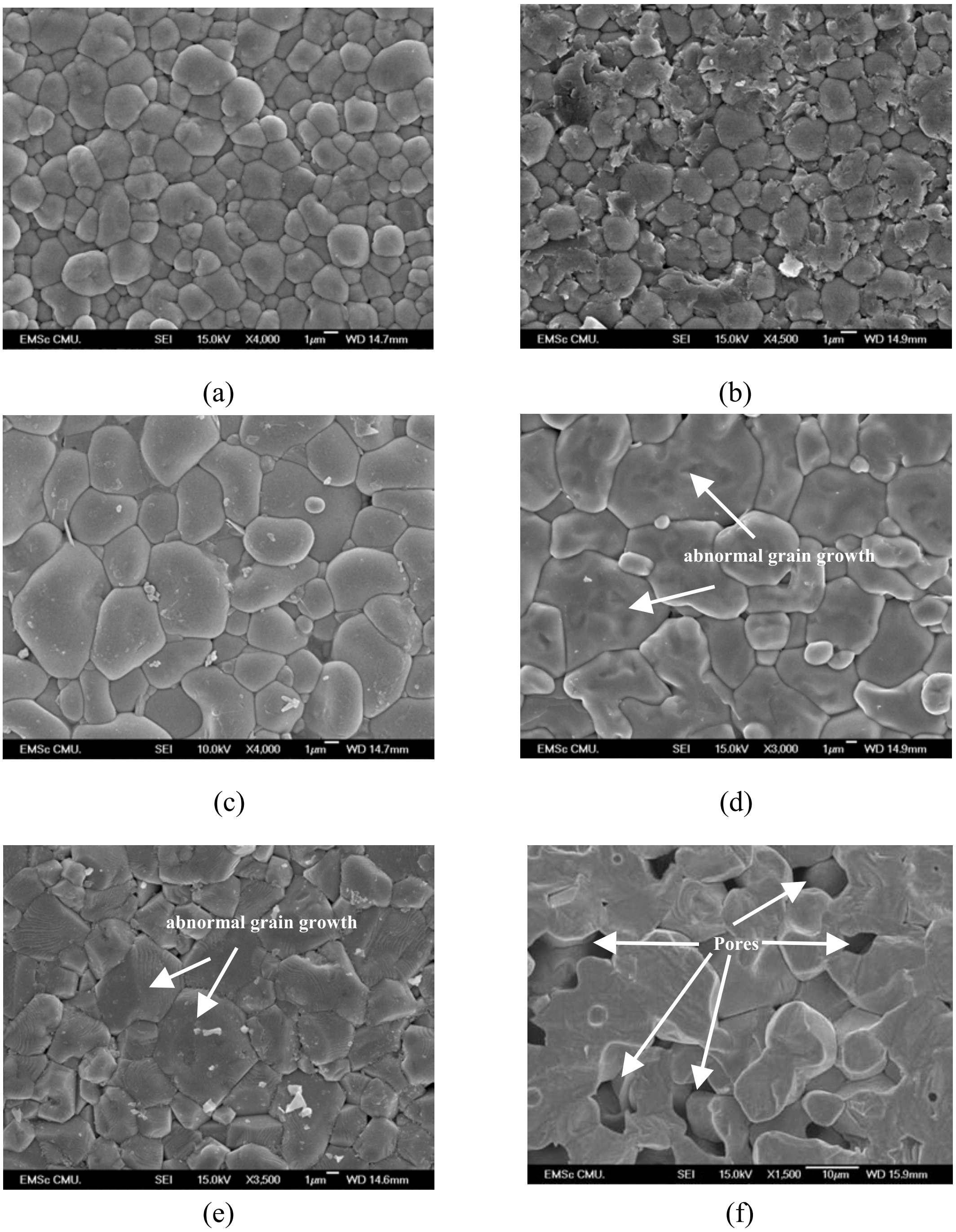

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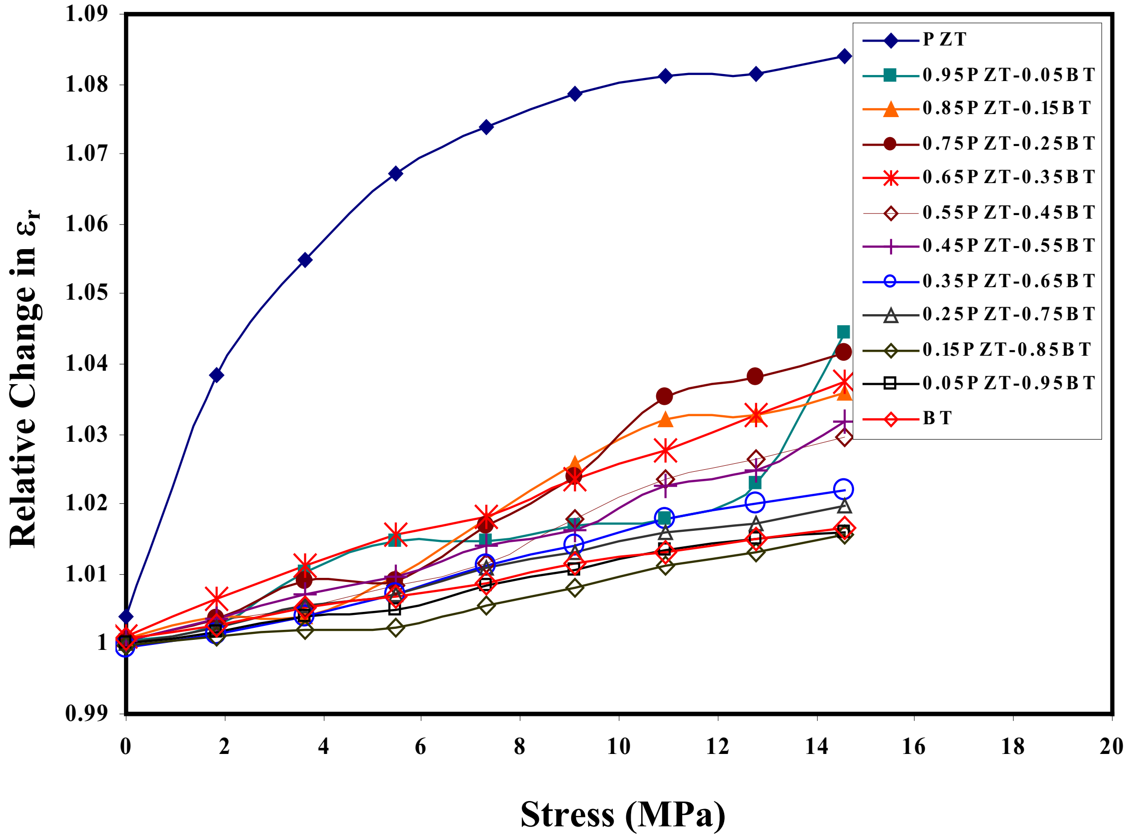

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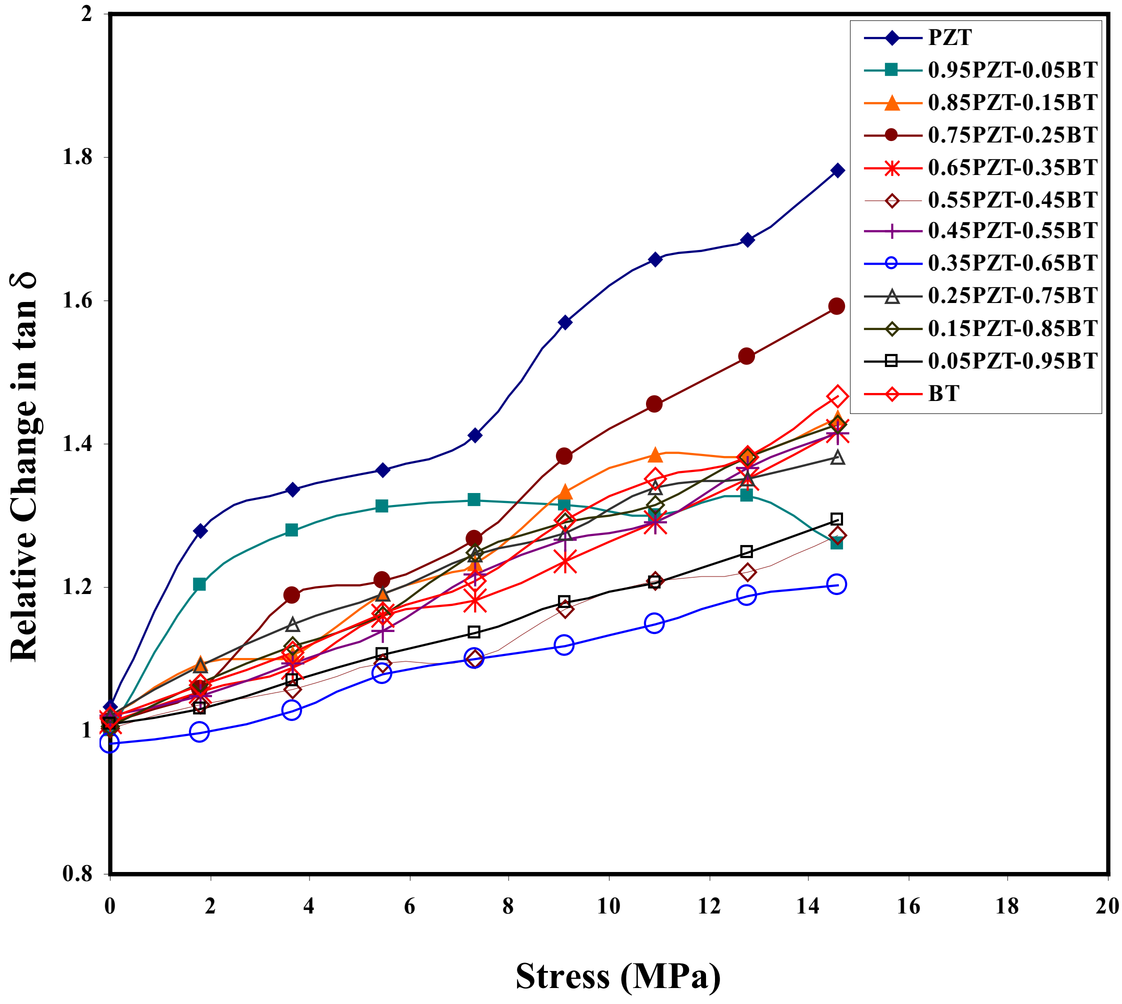

Fig. 5. Relative changes of dielectric loss tangent (tan δ) as a function of compressive pre-stress for (*1-x*)PZT-(*x*)BT ceramics.

Dielectric Properties of Complex Perovskite PZBT-PMNT Ceramic Under Compressive Stress

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ABSTRACT

Effects of compressive stress on the dielectric properties of complex perovskite PZBT-PMNT ceramic were investigated. The dielectric properties measured under stress-free condition showed a composite nature with two distinct temperatures of dielectric maximum associated with PZBT and PMNT. The dielectric properties under the compressive stress were observed at stress levels up to 230 MPa using a homebuilt compressometer. The results clearly showed that the superimposed compression load significantly reduced both the dielectric constant and the dielectric loss tangent in every measuring frequency. The change of the dielectric constant with stress was attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions, while the clamping of the domain walls contributed to the change in dielectric loss tangent with stress. Finally, the stress induced decrease in switchable part of spontaneous polarization was seen as a cause for a large drop of the dielectric constant after a stress cycle.

Keywords: Dielectric Properties, PZBT-PMNT, Stress

1. INTRODUCTION

Complex perovskite ferroelectric ceramics have been studied extensively because they can be applied in several micro- and nano-electronic devices such as multilayer capacitors, microactuators and miniaturized transducers [1-5]. Among them, barium titanate ($BaTiO_3$ or BT), lead titanate ($PbTiO_3$ or PT), lead zirconate titanate ($Pb(Zr_{1-x}Ti_x)O_3$ or PZT), lead magnesium niobate ($Pb(Mg_{1/3}Nb_{2/3})O_3$ or PMN) ceramics and a variety of their solid solutions have been investigated extensively and continuously since the late 1940s [1,3,4]. Two of the most studied ferroelectric compounds, PZT and BT ceramics are representative perovskite piezoelectric and ferroelectric prototypes, respectively, because of their excellent electrical properties. While BT has a high dielectric constant with a relatively low Curie temperature (T_C) (~120 ºC), PZT has a higher T_C of 390 ºC which allows PZT-based piezoelectric devices to be operated at relatively higher temperatures [2,4]. Although BT ceramic has better mechanical properties than PZT, the sintering temperature is also higher [1,3,6]. Thus, mixing PZT with BT is expected to decrease the sintering temperature of BT-based ceramics while maintaining excellent dielectric properties. Our earlier studies have already revealed the optimized electrical properties in 0.5PZT-0.5BT ceramic with $\varepsilon_r \sim 4000$ at T_C of 160 ºC [7,8].

Another family of ferroelectric materials which are of great interest due to their high polarizabilities is lead-based relaxor ferroelectrics. PMN has very good dielectric properties [2,3,5]. However, the temperature related to the maximum dielectric constant (T_{max}) of PMN is very low (~ -10°C) [5]. Therefore, PT is systematically added to PMN to enhance the dielectric properties of PMN (as well as increasing T_{max}). In particular, 0.9PMN-0.1PT ceramics, which have $T_{max} \sim 40$ °C and $\varepsilon_{r,max} > 20000$, have been a subject of many investigations and, more importantly, have been widely applied for capacitor, actuator and transducer applications [9-12]. However, since there have always been a need to obtain ceramic with board dielectric peak and maximum dielectric constant for various applications, especially in capacitive components [2-5], and with the complimentary characteristics between 0.5PZT-0.5BT and 0.9PMN-0.1PT ceramics, it is expected that excellent dielectric properties can be obtained from a mixture composition of the two ceramics. It is, therefore, one of the aims of this study to explore the dielectric properties of the mixture composition of 0.5PZT-0.5BT and 0.9PMN-0.1PT.

In most applications, however, ceramics are often subjected to mechanical loading, either deliberately in the design of the device itself or because the device is used to change shapes as in many smart structure applications or the device is used under environmental stresses [13-15]. A prior knowledge of how the materials behave under different load conditions is therefore very crucial for proper design of a device and for suitable selection of materials for a specific application. Therefore, it is very important to determine the properties of the materials as a function of applied stress. Previous investigations on the stress-dependence dielectric and electrical properties of many ceramic systems, such as PZT, PMN-PZT, and PMN-PT have clearly emphasized the importance of the subject [15-22]. Therefore, the major aim of this study is to investigate the influences of the compressive stress on the dielectric properties of complex perovskite PZBT-PMNT ceramic.

2. EXPERIMENTAL DETAILS

The ceramic composition with a formula $[0.5Pb(Zr_{0.52}Ti_{0.48})O_3–0.5BaTiO_3]$-$[0.9Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.1PbTiO_3]$ (abbreviated as PZBT-PMNT hereafter) was selected for this study as outlined above. The PZBT-PMNT powders were prepared from $0.5Pb(Zr_{0.52}Ti_{0.48})O_3$–$0.5BaTiO_3$ (abbreviated as PZBT hereafter) and $0.9Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.1PbTiO_3$ (abbreviated as PMNT hereafter) starting powders via a simple mixed-oxide method [7,23]. The detailed descriptions of the PZBT and PMNT powders processing and characterization were presented elsewhere and will not be discussed here [7,8,23]. The PZBT-PMNT ceramics were then fabricated via the same mixed-oxide method. After mixing the powders by ball-milling method and drying process, the mixed powders were pressed hydraulically to form disc-shaped pellets 10 mm in diameter and 2 mm thick, with 3 wt.% polyvinyl alcohol as a binder. The pellets were placed on the alumina powder-bed inside alumina crucible and surrounded with atmosphere powders of the same composition. Finally, the pellets were sintered at 1250 °C for 2 h.

For dielectric property characterizations, the sintered samples were lapped to obtain parallel faces disc-shaped specimens with diameter of 8 mm and thickness of 1 mm, and the faces were then coated with silver paint as electrodes. The samples were then heat-treated at 750 °C for 12 min to ensure the contact between the electrodes and the ceramic surfaces. The samples were subsequently poled in a silicone oil bath at a temperature of 120 °C by applying a dc field of 20 kV/cm for 30 min and then field-cooled to room temperature. The dielectric properties of the sintered ceramics were studied under stress-free condition as functions of both temperature and frequency with an automated dielectric measurement system. The capacitance and the dielectric loss tangent were determined over the temperature range of 25 and 400 °C with the frequency ranging from 1 kHz to 5 MHz. The measurements were carried out on cooling continuously at the rate of 3 °C/min. To study the effects of the compressive stress on the dielectric properties, the compressometer was constructed [20,24]. The dielectric properties were measured by LCR-meter (Instrek LCR-821). The room temperature (25 °C) capacitance and the dielectric loss tangent were obtained at frequency range 10 to 200 kHz under the compressive stress levels up to 230 MPa. The dielectric constant was then calculated from a parallel-plate capacitor equation, e.g. $\varepsilon_r = Cd/\varepsilon_0 A$, where C is the capacitance of the specimens, d and A are, respectively, the thickness and the area of the electrode, and ε_0 is the dielectric permittivity of vacuum (8.854×10^{-12} F/m).

3. RESULTS AND DISCUSSION

The stress-free dielectric properties, e.g. dielectric constant (ε_r) and tan δ, as functions of both temperature and frequency are plotted in Fig. 1. It is clearly seen that there are two distinct dielectric anomalies. The first dielectric peak at lower temperature region with higher dielectric constant is associated with the PMNT component, which has T_{max} near 40 ºC [12]. A strong dielectric dispersion below T_{max} also indicates a relaxor ferroelectric behavior. In this case, the temperatures of maximum dielectric constant and dielectric loss tangent are shifted to higher temperature with increasing frequency. The maximum value of the dielectric constant decreases with increasing frequency, while that of the dielectric loss tangent increases. The dielectric properties then become frequency independence above the transition temperature [25,26]. Even though these descriptions are not clearly seen on the first dielectric peak because of the appearance of the second dielectric peak, it could still be said that the relaxor behavior exists. The second dielectric peak occurs over temperature range 150-200 ºC, which should be associated with the PZBT component with T_C near 162 ºC [7,8]. A diffuse phase transition with small frequency dispersion of the dielectric maxima is also observed within this second dielectric peak, similar to earlier reported observation for the PZBT component [8]. These observations of the two distinct dielectric peaks should indicate the composite nature of the PZBT-PMNT ceramic. Moreover, it should also be noted that significant increase in the dielectric constant dielectric loss at high temperatures is a result of thermally activated space charge conduction [25,26].

The fractional changes of the dielectric properties of the PZBT-PMNT ceramic under the compressive stress during loading and unloading are shown in Figs. 2 and 3. There is a significant change of the dielectric properties of the ceramic with increasing stress from 0 to 230 MPa and returning to stress-free condition. As seen in Fig. 2, the dielectric constant decreases monotonically with increasing the compressive stress, then increases only slightly when the compressive stress is gradually removed. The changing of the dielectric constant with increasing and decreasing the applied stress does not follow the same path. In every frequency, the dielectric constant with increasing the compressive stress is larger in value than that with decreasing stress. It is also of interest to see that the stress-free dielectric constant value decreases significantly after a stress cycle. It can be seen very clearly that the dielectric constant decreases as much as 25% at the maximum applied stress and only returns to slightly less than 85% of its original value when the stress is removed. As displayed in Fig. 3, though following a similar trend, the change in the dielectric loss tangent value is less significant, as it only decreases about 10-20% at the maximum stress and almost returns to its original value after a stress cycle. Similar observations have been reported for other ceramic systems, such as BT, PZT, PMN, PMN-PT and PMN-PZT [17,20,22,27-28]. It should also be noted that since the dielectric constant of the sample was measured through the capacitance, there is a change of sample capacitance due to the geometrical deformation under stress. The variation of the sample dielectric constant ($\Delta\varepsilon_r$) can be expressed as $\varepsilon_r * X * ((1+2\upsilon)/E)$, where X is the applied stress, υ is the Poisson's ratio, and E is the Young's modulus [29,30]. By applying the estimated values of $\upsilon \sim 0.3$ and $E \sim 100$ GPa for the ceramic [15,16,18,22,31] and $\varepsilon_r \sim 4000$, it can be estimated that at the maximum stress of 230 MPa, the variation of the sample dielectric constant due to the geometrical deformation is < 0.5%. Therefore, this variation should not be an important factor in the variation of the dielectric constant under stress observed in Fig. 2.

To understand these experimental results, at least qualitatively, various effects have to be considered. Normally, the properties of ferroelectric materials are derived from both the intrinsic contribution of domains and extrinsic contributions of re-polarization and growth of micro-polar regions [16,17,20,22,27,28]. When a compressive stress is applied to the ferroelectric materials, the domain structure in the materials will change to maintain the domain energy at a minimum; during this process some of the domains engulf other domains or change shape irreversibly. Under the applied stress, the domain structure of ferroelectric ceramics may undergo domain switching through non-180° domain walls, de-aging, clamping of domain walls, and stress induced decrease in switchable part of spontaneous polarization [21,27,31,32,33].

The situation for the PZBT-PMNT system is quite complex because this system is a mixing between the relaxor ferroelectric PMNT and the normal ferroelectric PZBT. Therefore, there is a competing mechanism between the two types of materials, depending upon temperature. Since the stress experiment was carried out at room temperature (~25 °C) which is just below the T_{max} of PMNT (~40 °C) [12], the dielectric properties of the PZBT-PMNT ceramic undet stress should be dominated by the PMNT contribution. Therefore, the experimental observations, which show decreases in both dielectric constant and dielectric loss tangent with increasing stress, can be attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. Since the dielectric response of both contributions is affected by the applied stress in an opposite way, the behavior of the PZBT-PMNT ceramic depends on the ratio between the micro-polar region and the non-polar matrix, in this case the non-polar matrix still dominates [27,28]. Hence, the dielectric responses of the PZBT-PMNT ceramic are observed to decrease significantly with increasing the compressive stress, as seen in Figs. 2 and 3. More interestingly, it is worth noting that our earlier investigations on the stress-dependent dielectric properties of the two members, i.e. PZBT [32] and PMNT [22], revealed that under a similar loading condition the dielectric constant of the PMNT ceramic decreased nearly 70%, whereas the PZBT ceramic showed approximately 5-10% increase in the dielectric constant. Clearly, the apparent suppression of the dielectric constant change with stress observed in this current study supports the composite nature of the PZBT-PMNT ceramic, as also revealed by the free-stress dielectric properties measurements shown in Fig. 1.

The cause of the stress dependence of the dielectric loss tangent is a little more straightforward than that of the dielectric constant. As depicted in Fig. 3, the clamping of the domain walls under the compressive stress results in a decrease of domain wall mobility and reduces the dielectric loss tangent [22,27]. This is a reversible effect with the domain wall mobility returning to near the original values when the applied stress is removed, as seen in Fig. 3 that the dielectric loss tangents return to near their original values after a stress cycle. In addition, a significant decrease in the dielectric constant after a full cycle of stress application has been observed, and attributed to the stress induced decrease in switchable part of spontaneous polarization at high stress [21,31].

4. CONCLUSIONS

The dielectric properties of complex perovskite PZBT-PMNT ceramic were investigated in this study. The dielectric properties measured under stress-free condition showed two distinct dielectric peaks associated with PZBT and PMNT. The dielectric properties under the compressive stress were observed using a homebuilt compressometer. The results clearly showed that the superimposed compression load significantly reduced both the dielectric constant and the dielectric loss tangent in every measuring frequency. The change of the dielectric properties with stress was attributed to competing influences of the intrinsic contribution of non-polar matrix and the extrinsic contribution of re-polarization and growth of micro-polar regions. Other contributions include the clamping of the domain walls and the stress induced decrease in switchable part of spontaneous polarization. This study clearly indicated that the complex perovskite PZBT-PMNT ceramic possessed a composite nature, as confirmed by the dielectric measurements.

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List of Figure Captions

Fig. 1. Temperature and frequency dependences of dielectric properties of PZBT-PMNT ceramic under stress-free condition (solid circle indicates dielectric peak associated with PMNT, and dotted circle indicates dielectric peak associated with PZBT).

Fig. 2. Relative changes of dielectric constant (ε_r) as a function of compressive stress for PZBT-PMNT ceramic.

Fig. 3. Relative changes of dielectric loss tangent (tan δ) as a function of compressive stress for PZBT-PMNT ceramic.

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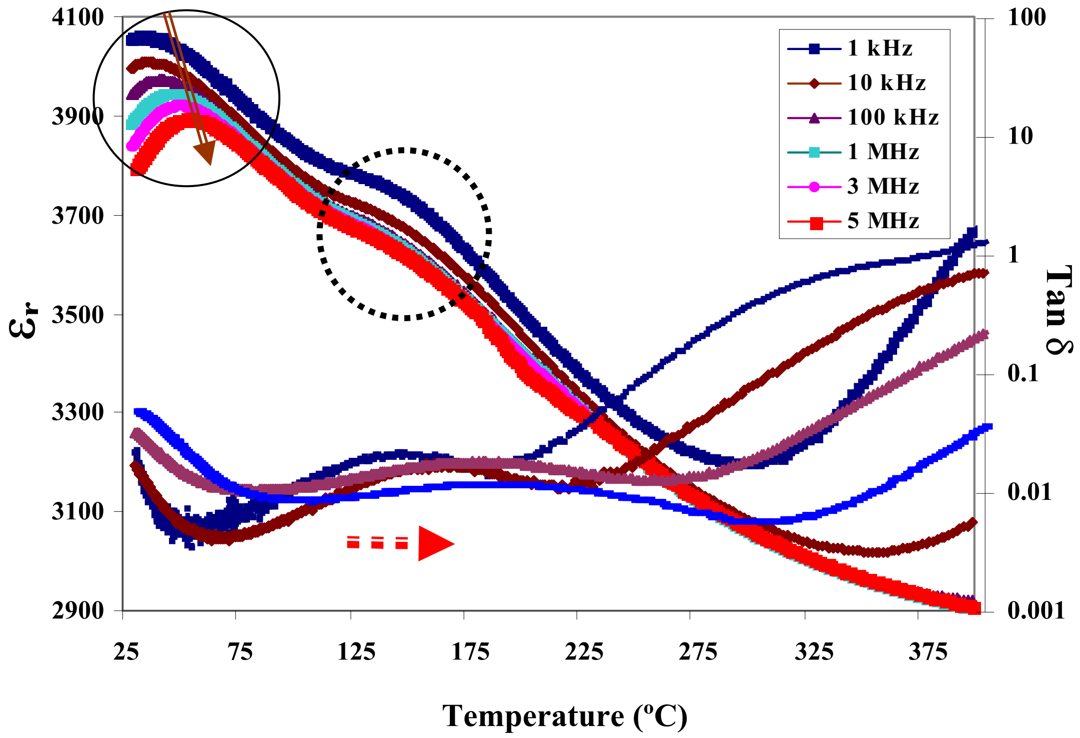

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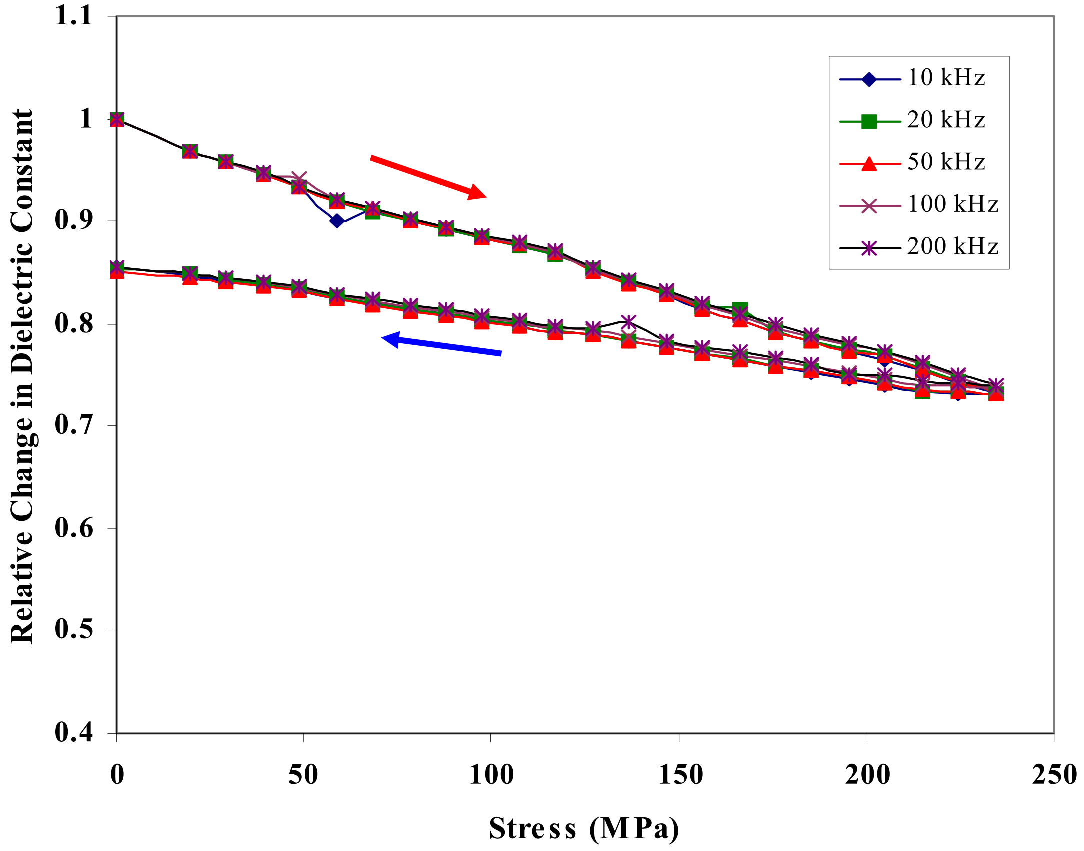

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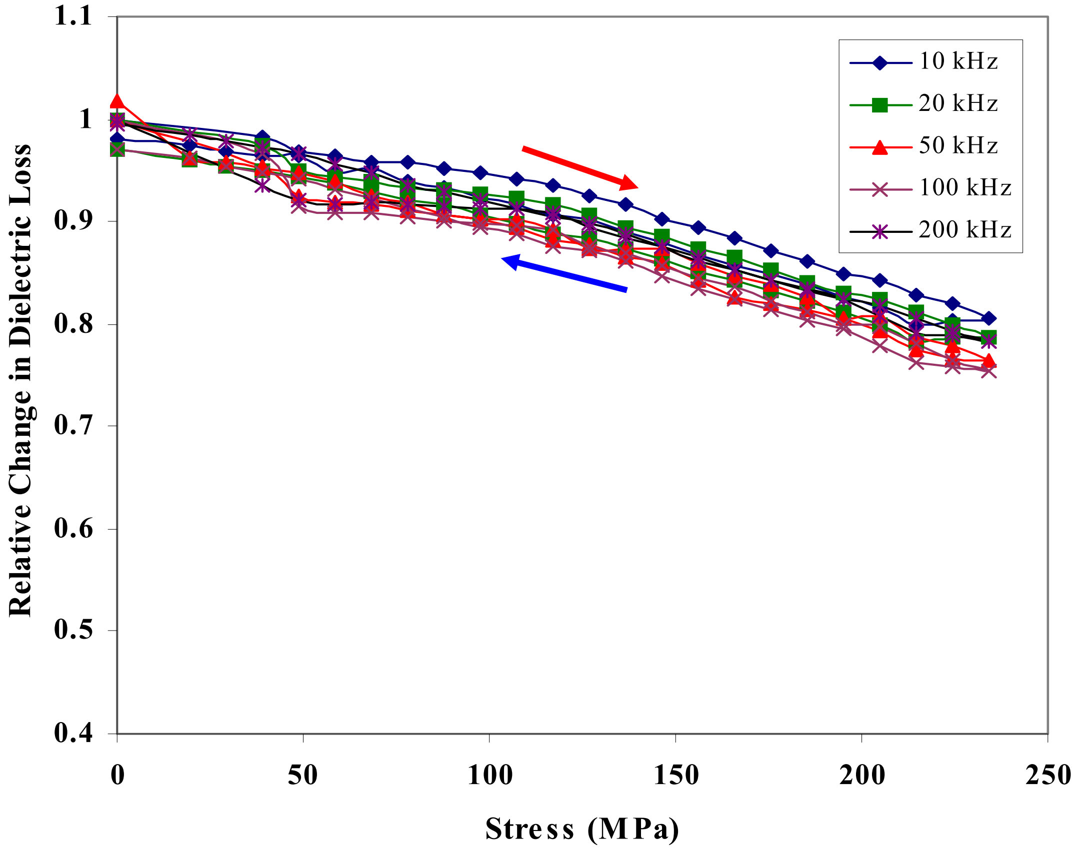

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